ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

Winter 1965-1966

● VOLUME 3

● NUMBER 2

TECHNICAL PROGRESS REVIEWS

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Argonne National Laboratory

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ERRATUM

In Table 2 on page 6 of *Isotopes and Radiation Technology*, Vol. 3, No. 1, the half-lives given in "months" should be in "minutes."

Issued quarterly by the U. S. Atomic Energy Commission. Use of funds for printing this publication approved by the Director of the Bureau of the Budget on December 23, 1965.

Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Medicine
Safety
Isotopes Around the World
Miscellaneous Items of Interest

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R. E. GREENE, and F. E. McKINNEY, *Editors*
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Section I

Isotopes and Radiation Technology

Isotope Production and Development

Production of Carrier-Free Radioactive Isotopes with the University of Pittsburgh Cyclotron

By A. J. Allen and B. L. Cohen*

Since the nuclear physics research program at the University of Pittsburgh is being transferred to a new tandem Van de Graaff facility, the 47-in. cyclotron at the University is now available for production of carrier-free radioisotopes.

This cyclotron is used principally for accelerating deuterons to 15 Mev, and isotopes are produced most efficiently by intercepting the circulating beam with an internal target inserted into the vacuum system at the point labeled "internal target position" in Fig. I-1. Beam currents up to 200 μa are available at this position, and currents of about 10 μa are available at the "external target position." (The apparatus in Fig. I-1 between the bellows and auxiliary magnet has since been removed.) The beam passes through a window into the air, so the target is not in the cyclotron vacuum. This allows the use of gas targets or of targets whose materials cannot be adequately packaged and cooled without affecting the vacuum.

A typical internal target, shown in Fig. I-2, is a cup that is sealed by O-rings to a shaft containing cooling-water entries and exits. The cup is thus cooled from the inside by a high-velocity stream of chilled, demineralized water. The cyclotron beam strikes the beveled edge of the cup at a glancing angle, as shown in Fig. I-2. A motor in the floor (Fig. I-1) rotates the cup by a system of V-belts and pulleys to spread the area struck by the beam over the entire beveled surface of the cup. The heating produced by the beam is so localized and intense that, if the rotation of the target stops, a hole is instantly burned in the cup. To guard against this, a safety interlock shuts off the cyclotron oscillator if the target stops rotating.

A list of some typical radioisotopes and their estimated production rates on the Pittsburgh cyclotron is given in Table I-1, which shows that the most efficient production reaction is $(d,2n)$. However, isotopes produced by this reaction can generally be produced more efficiently in the Oak Ridge National Laboratory (ORNL) cyclotron by (p,n) reactions; the higher energies and larger currents in that machine give it an order of magnitude higher production rate. This is somewhat offset by a larger hourly charge, but obtaining these isotopes from ORNL will still usually be better. There are several exceptions to this. One exception is when external beams are used—in making ^{84}Rb, for example—since the Pittsburgh external beam current is about the same as that at ORNL. (A gas target has been developed for use with the external beam.) Another exception is when only a relatively small amount of the radioisotope is needed, since the minimum charge at Pittsburgh is far less than at ORNL. The Pittsburgh cyclotron will also be useful for $(d,2n)$ reactions when the ORNL cyclotron is shut down for repairs and when half-lives are so short that delivery distance and transportation connections are significant.

There are several isotopes that can be produced by deuterons more efficiently than by protons. The most important example is ^{22}Na, by the $Mg(d,\alpha)$ reaction. A suitable magnesium

*University of Pittsburgh, Pittsburgh, Pa.

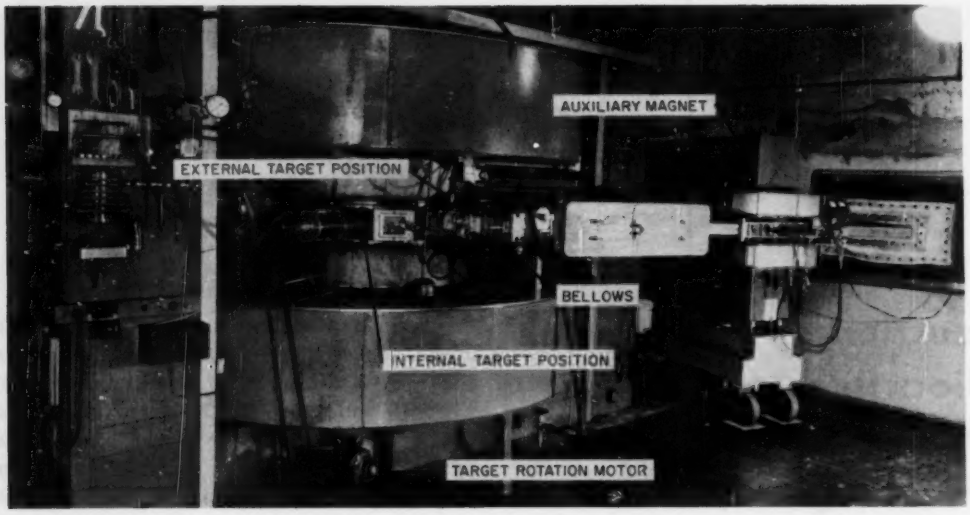

Fig. I-1 University of Pittsburgh 47-in. cyclotron showing irradiation facilities for carrier-free radioisotope production.

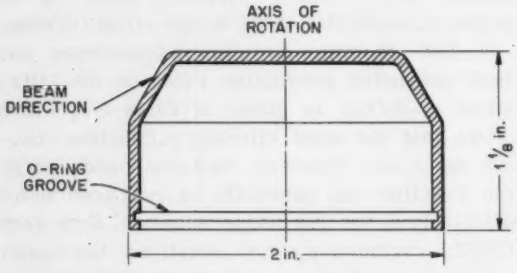

Fig. I-2 Standard copper-cup target used on University of Pittsburgh 47-in. cyclotron.

Table I-1 EXAMPLES OF ISOTOPE PRODUCTION RATES IN THE UNIVERSITY OF PITTSBURGH CYCLOTRON

Isotope	Half-life	Target material	Reaction	Estimated production rate, mc/hr	Target* charge, $	Notes
^{7}Be	53 days	LiF	$(d,2n)$	0.4	5	External beam
^{22}Na	2.6 years	Mg	(d,α)	0.3†	50	
^{46}Sc	84 days	Ti	(d,α)	0.5	35	
^{51}Cr	27 days	V	$(d,2n)$	17.0	30	
^{52}Mn	5.7 days	Cr	$(d,2n)$	20.0†	25	
^{54}Mn	314 days	Fe	(d,α)	0.2†	25	
^{55}Fe	2.7 years	Mn	$(d,2n)$	0.5†	60	
^{57}Co	267 days	Fe	(d,n)	1.5†	25	
^{58}Co	71 days	Fe	(d,n)	0.2	25	
^{59}Fe	45 days	Co	$(d,2p)$	0.02	20	
^{65}Zn	245 days	Cu	$(d,2n)$	0.7	10	
^{74}As	18 days	Ge	$(d,2n)$	10.0	85	
^{84}Rb	33 days	Kr	$(d,2n)$	0.4	50‡	External beam, gas target
^{85}Sr	64 days	Rb–salt	$(d,2n)$	0.2	5	External beam
^{109}Cd	1.3 years	Ag	$(d,2n)$	0.6	25	
^{197}Hg	65 hr	Au	$(d,2n)$	150.0	40	
^{206}Bi	6.2 days	Pb	$(d,2n)$	12.0	35	

*Targets are provided by Fodor Targets Company by plating various materials on the beveled surface of a standard copper-cup target (Fig. I-2).

†Checked experimentally by I. Gruverman (private communication).

‡Rental fee. A target can be purchased for $300 and be used repeatedly.

target for ^{22}Na production has been developed with the assistance of S. L. Couling of Dow Metal Products Company, Metallurgical Laboratory, and the availability of the Pittsburgh cyclotron should substantially reduce the price of this isotope. Other isotopes produced by (d,α), (d,n), and $(d,2p)$ reactions are listed in Table I-1; these reactions give products that generally cannot be produced efficiently with protons.

The isotope production program as outlined will be a self-sustaining, nonprofit enterprise. For now, the operating cost has been set at $30 per hour. Production rates listed in Table I-1 are not guaranteed, but adjustments in the charges will be made for any large discrepancies. In addition, the purchaser is expected to pay for 1 hr of setup time, for target fabrication (Table I-1), and for shipping. Customers have the option of preparing their own targets by plating on copper cups that will be supplied at a small cost. Since there are no facilities available for radiochemical processing, all targets will be shipped as bombarded.

Inquiries or orders, addressed to B. L. Cohen, are invited.

Section

II

Isotopes and Radiation Technology

Isotope Technology Development

A symposium on Low-Energy X- and Gamma Sources and Applications held at Illinois Institute of Technology Research Institute (IITRI) on Oct. 21–22, 1964, is summarized in this section. These papers, which gave information on source development and medical uses of isotopes as well as on isotope technology, are all discussed here rather than in the appropriate section so that the entire summary will appear in one place.

A review of uses of radioisotopes in the development of petroleum and natural-gas reservoirs, given by F. E. Armstrong and W. D. Howell at the Third Symposium on the Development of Petroleum Resources of Asia and the Far East in Tokyo in November 1965, is also presented.

Low-Energy X- and Gamma Sources and Applications

The Symposium on Low-Energy X- and Gamma Sources and Applications held at Illinois Institute of Technology Research Institute on Oct. 21–22, 1964, is summarized. Papers were given there on (1) preparation and evaluation of low-energy X and gamma sources and on the use of such sources for (2) chemical analysis by X-ray fluorescence and absorption, (3) industrial gaging, (4) industrial radiography, and (5) medical radiography, bone analysis, and organ visualization.

The Proceedings of the Symposium on Low-Energy X- and Gamma Sources and Applications held at the Illinois Institute of Technology Research Institute (IITRI) on Oct. 21–22, 1964, have now been published.[1] Although a preliminary program and a brief synopsis of this conference have already appeared in *Isotopes and Radiation Technology*,[2] more extensive coverage of the conference proceedings is given here because this conference provided an excellent state-of-the-art summary of this important field of isotope technology.

This review is presented in the following broad subject categories: (1) source preparation and evaluation, (2) chemical analysis by X-ray fluorescence and absorption, (3) industrial gaging, (4) industrial radiography, and (5) medical radiography, bone analysis, and organ visualization by isotopic techniques. The papers are referred to by the numbers given in Table II-1.

Source Preparation and Evaluation

Most of the papers presented at the symposium gave some information on the preparation and properties of sources of low-energy gamma and X rays. These sources may, for convenience, be classified according to the method of producing the radiation: (1) direct radiation production, which includes gamma rays and K-capture or internal-conversion X rays; (2) secondary radiation production resulting from the interaction of primary nuclear particles with matter, which includes bremsstrahlung and characteristic X rays produced by beta particles and characteristic X rays produced by alpha particles. Sources of both types were discussed in earlier issues of *Isotopes and Radiation Technology*.[3,4]

LOW-ENERGY GAMMA EMITTERS

Cameron and Florkowski (paper 12) summarized the low-energy gamma-emitting isotopes that are now in general use (Table II-2). Case and Pressly (paper 3) presented results of some work at Oak Ridge National Laboratory (ORNL) on ^{151}Sm, ^{148}Pm, ^{79}Kr, and ^{131m}Xe. Samarium-151, which decays by 0.075-Mev-

Table II-1 TITLES OF PAPERS PRESENTED AT SYMPOSIUM ON LOW-ENERGY X- AND GAMMA SOURCES AND APPLICATIONS

Paper No.	Title	Author	Author's affiliation	Pages in proceedings
1	Background and Purpose of the Symposium	J. W. Hitch	USAEC, Washington*	1–3
2	Study, Design, and Applications of Beta-Excited X-Ray Sources	J. J. Ezop and T. G. Stinchcomb	Illinois Institute of Technology Research Institute, Chicago, Ill.	4–37
3	Recent Developments in Production of Low-Energy Gamma- and X-Ray Emitters	F. N. Case and R. S. Pressly	Oak Ridge National Laboratory,* Oak Ridge, Tenn.	38–50
4	X-Ray Absorptiometry with Beta-Stimulated X-Ray Sources and Progress Report on Compilation of Beta-Excited Spectra	L. E. Preuss, H. Collins, and J. Kann	Edsel B. Ford Institute for Medical Research,* Detroit, Mich.	51–71
5	Analysis and Measurement of Materials with Low-Energy Radiation	C. O. Badgett	Industrial Nucleonics Corp., Columbus, Ohio	72–90
6	Bremsstrahlung from ^{90}Sr-^{90}Y in Thin Absorbers	T. S. Bustard and J. Silverman	Hittman Associates, Inc., Baltimore, Md.; University of Maryland, College Park	91–98
7	External Bremsstrahlung Produced in Thin Foils by ^{147}Pm	V. C. Truscello and J. Silverman	Martin Company, Martin Marietta Corporation, Baltimore, Md.; University of Maryland, College Park	99–106
8	Heavy Elements Content Measurement by Means of Gamma-Excited X-Ray Fluorescence	P. Martinelli and P. Blanquet	CEA, France; Société Minerais et Métaux, France	107–123
9	Americium-241 as a Nondestructive Testing Tool for the Aircraft and Space Industry	G. Locher	Western Radiation Laboratory, Los Angeles, Calif.	124–137
10	Preliminary Results of X-Ray Fluorescence Analysis in the Energy Range 1 to 5 Kev	T. C. Martin, K. R. Blake, and I. L. Morgan	Texas Nuclear Corporation,* Austin, Tex.	138–153
11	Developments in a Portable X-Ray Fluorescence Instrument using Radioisotope Excitation Sources	J. O. Karttunen and D. J. Henderson	Argonne National Laboratory, Argonne, Ill.	154–181
12	Radioisotope Sources of Low-Energy Electromagnetic Radiation and Their Use in Analysis and Measurement of Coating Thickness	J. F. Cameron and T. Florkowski	IAEA, Kaerntnerring, Vienna I, Austria	182–205
13	Ore and Coal Analysis using Radioisotope Techniques	J. R. Rhodes	UKAEA, Wantage Research Laboratory, Wantage, Berkshire, England	206–221
14	Industrial Analysis of Promethium-147 X Rays for Radiography and Imaging Readout	E. W. Coleman, T. C. Furnas, J. Ball, M. R. Rosumny, and D. W. Beard	Picker X-Ray Corp., Cleveland, Ohio	222–230
	Round-Table Discussion			231–238
15	Medical Applications of Low-Energy X-Ray and Gamma Radiation Sources	H. N. Wagner, Jr.	Johns Hopkins Medical Institutions, Baltimore, Md.	239–242
16	Use of Iodine-125 for Intraoral X Radiography	R. Söremark	Harvard University, Boston, Mass.	243–256
17	Radioisotopic Device for Measuring Bone Mineral	L. H. Lanzl and N. Strandjord	University of Chicago and Argonne Cancer Research Hospital, Chicago, Ill.	257–276
18	Body Composition Determination by Differential Absorption of Monochromatic X Rays	J. A. Sorenson and J. R. Cameron	University of Wisconsin, Madison	277–289
19	In Vivo Measurement of Bone Mineral Content by a Transmission Technique	L. S. Goodman and B. Levin	Argonne National Laboratory, Argonne, Ill.; Michael Reese Hospital, Chicago, Ill.	290–294
20	An Image Tube Scintillation Camera	M. M. Ter-Pogossian	Edward Mallinckrodt Institute of Radiology, Washington University, St. Louis, Mo.	295–308
21	Portable Devices for Isotope Chest Radiography and Other Applications	F. L. Green	Viso Corporation, Detroit, Mich.	309–328
22	Recent Research Developments at Polaroid Corporation and Their Application to Low Energy Radiography	H. J. Frede	Polaroid X-Ray Research Laboratories, Waltham, Mass.	329–338
23	Proton-Excited X Rays and Applications	A. A. Sterk	American Machine & Foundry Co., Alexandria, Va.	339–352
24	Generation and Practical Use of Monoenergetic X Rays from Alpha-Emitting Isotopes	B. Sellers and C. A. Ziegler	Parametrics, Inc.,* Waltham, Mass.	353–373

*Work sponsored by Division of Isotopes Development.

Table II-2 GAMMA-RAY, K-CAPTURE, AND INTERNAL-CONVERSION SOURCES*

Radioisotope	Half-life	Decay			γ energy, kev; and (abundance)	Internal-conversion factor, %	X-ray energy, kev
		Type	Energy, kev	Abundance, %			
^{55}Fe	2.7 years	E.C.		100	–	–	5.9
^{109}Cd	470 days	E.C.		100	88 (4%)	96	22
^{125}I	57.4 days	E.C.		100	35.4 (7%)	93	27
^{241}Am	470 years	α	5.44 Mev	13	27 (2.8%)		
			5.48 Mev	85	33 (0.2%)		11–22
					60 (40%)		
					Others weaker		
^{153}Gd	236 days	E.C.		100	70 (2%)	9	
					97 (21%)	8	41
					103 (25%)	35	
^{170}Tm	127 days	β^-	880	22	84 (3%)	19	52
			970	78			7
^{155}Eu	1.7 years	β^-	154	80	87		
			243	20	105		
			Others weaker		Others weaker		43
^{145}Sm	1.0 year	E.C.		100	61 (14%)	78	39
^{57}Co	270 days	E.C.		100	14 (6%)	83	
					122 (88%)	1	6.4
					136 (10%)	1	

*From paper 12.

beta and 0.021-Mev-gamma emissions and has a half-life of 93 years, is formed by fission and is separated along with inactive samarium in the ion-exchange purification of ^{147}Pm. Promethium-148 is produced by the (n,γ) reaction on ^{147}Pm in a reactor. Two isomers, with 5.4- and 41.8-day half-lives, are produced. Krypton-79, which decays by 1.33- and 1.11-Mev-gamma emissions and has a 34.5-hr half-life, is produced by the ^{78}Kr$(n,\gamma)^{79}$Kr reaction on a target enriched to about 7% in ^{78}Kr. Neutron irradiation of ^{129}I (from fission) produces ^{131m}Xe, which decays by a 0.164-Mev-gamma emission and has a 12-day half-life. These reactions are

$$^{129}\mathrm{I}(n,\gamma)^{130}\mathrm{I} \xrightarrow[12.5\ \mathrm{hr}]{\beta^-} {}^{130}\mathrm{Xe}$$

$$^{130}\mathrm{Xe}(n,\gamma)^{131m}\mathrm{Xe}$$

CHARACTERISTIC X-RAY SOURCES

The production of characteristic X rays by excitation of targets with beta rays from ^{147}Pm, ^{90}Sr, ^{90}Y, ^{85}Kr, ^{99}Tc, and ^{3}H was discussed by Ezop and Stinchcomb (paper 2), Case and Pressly (paper 3), Preuss, Collins, and Kann (paper 4), and Cameron and Florkowski (paper 12). Since much of this work was reviewed in a recent article in *Isotopes and Radiation Technology*,[4] it is not discussed here.

Sellers and Ziegler (paper 24) reported studies of the excitation of characteristic X rays with a 0.69-mc ^{210}Po alpha source. Sources of this type are essentially monoenergetic. Almost no continuous radiation, or bremsstrahlung, is generated. Although the "yield" or number of X rays produced per alpha particle is smaller than that obtained with equivalent-energy beta rays, usable X-ray fluxes can be generated with millicurie alpha sources. The absence of a significant amount of high-energy penetrating radiation allows high source strength to be used safely with much less shielding than that required by beta-ray-excited sources.

The only information presented at the meeting on a machine source of radiation was that by Sterk (paper 23), who described the production of characteristic X rays by proton excitation. His basic apparatus (Fig. II-1) is a plasma generator in which hydrogen gas under a pressure of about 10^{-2} torr is ionized by a radio-frequency generator. A magnet coil confines the region of maximum plasma density to the center of the glass envelope. When a d-c voltage of the appropriate polarity is applied, protons are extracted through a small orifice and then accelerated through a single-stage electrostatic lens, under a potential that is adjustable from 0 to 150 kv. The accelerated and focused proton beam collides with a target, producing X rays of wavelength determined by target composition. In the experiments reported to date, targets of aluminum, oxygen (LiOH), and carbon have been used, with production of monochromatic characteristic K lines of the respective elements.

The device described is being used as a source of monochromatic X rays for the align-

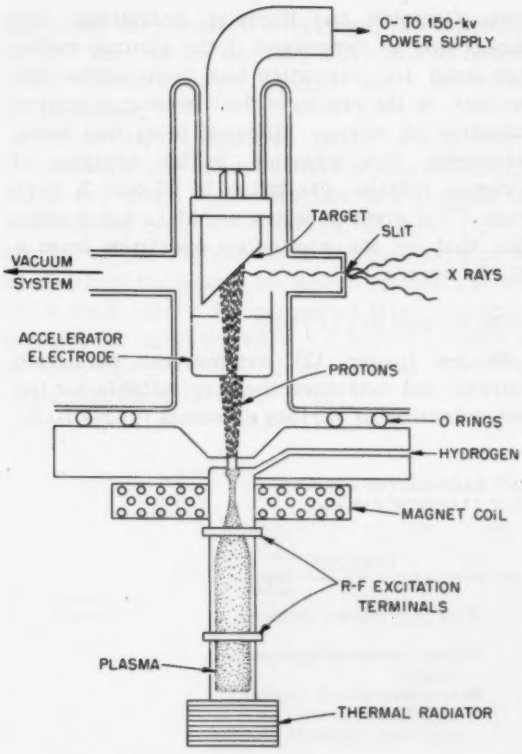

Fig. II-1 Proton-excited soft X-ray generator. (From paper 23.)

ment and calibration of X-ray spectrometers and will be used for X-ray absorption studies.

BREMSSTRAHLUNG

Bustard and Silverman (paper 6) and Truscello and Silverman (paper 7) reported on bremsstrahlung production in targets with atomic numbers from 13 to 73 and ^{147}Pm and $^{90}Sr-^{90}Y$ sources. The bremsstrahlung yield for both sources was found to be linearly dependent on the atomic number of the target material, as predicted by theory. For the $^{90}Sr-^{90}Y$ source, the yields can be calculated by the empirical equations

$$\text{Photon yield}/\beta = (1.50 \times 10^{-3})Z$$

$$\text{Energy yield, Mev}/\beta = (2.52 \times 10^{-4})Z$$

For the ^{147}Pm sources, the equations are

$$\text{Photon yield}/\beta = (3.7 \times 10^{-5})Z$$

$$\text{Energy yield, Mev}/\beta = (1.8 \times 10^{-6})Z$$

SOURCE EVALUATION

Coleman et al. (paper 14) recently completed an evaluation of a 100-curie ^{147}Pm source for use in medical and industrial radiography. As expected, the main difficulty with this source was the large focal spot; the radiographs lacked fine detail. The conclusion of this paper presented an interesting and different approach to the evaluation of isotope sources:

> Quite often in these meetings we talk about using isotopes and extol their advantages. Consider for a moment, however, something we have learned from experience in evaluating isotope equipment and also in trying to sell it to people. Assuming we have a suitable device for containing and controlling radiation, the following features are usually accepted as being true of isotope sources:
>
> 1. The radiation is immediately available.
> 2. The energy spectrum is unaffected by the operating history of the equipment.
> 3. For a given isotope in a fixed source geometry, the radiation intensity varies only in a uniform and predictable manner.
> 4. The character of nuclear sources makes possible nearly exact duplication of previous results.
>
> Now these same four properties are often given back to us as disadvantages. Listen to the same list of four characteristics as we often hear them:
>
> 1. The radiation is continuously produced and must be continuously controlled; deterioration or radioactive decay occurs even during periods when the radiation is not required.
> 2. The energy spectrum cannot be easily changed.
> 3. The radiation intensity is difficult to vary precisely; time and distance must be used as control variables.
> 4. Nuclear properties define the quality and quantity of radiation available and the possibilities are limited to a finite number of isotopes.
>
> Certainly in our thinking about isotopes, we ought to remember that some things that we see as advantages, other people see as disadvantages, and we ought to give consideration to both sides.

Analysis by X-Ray Fluorescence and Absorptiometry

Seven papers (papers 2, 4, 8, 10–13) described methods using isotopic sources for determining various elements by X-ray fluorescence and absorptiometry. Most of these papers are on X-ray fluorescence analysis, but there is also a short discussion of X-ray absorptiometry. Although numerous papers have been published on the use of radioisotopes as excitation sources in X-ray fluorescence analysis,[5–17] publication of work on the practical application of this method has just recently started. The papers presented at this conference, together with those presented at the In-

ternational Atomic Energy Agency Symposium on Radiochemical Methods of Analysis[18] held at Salzburg, Oct. 19–23, 1964, provide a good review of this subject. The papers of Martinelli and Blanquet (paper 8) and Cameron and Florkowski (paper 12) have excellent introductions to the subject of X-ray fluorescence analysis. This material is summarized here.

The use of radioisotope sources to excite characteristic X rays in X-ray fluorescence analysis (or X-ray spectrometry) necessitates a major change from the usual technique in which an X-ray tube is used for excitation.[19] The relatively low photon output (10^7 to 10^8 per second) of radioisotope sources prevents the use of conventional dispersion methods of energy selection of the excited X rays because of the intensity loss. However, nondispersive spectrometry with X-ray filters makes possible the isolation of a given characteristic X ray from neighboring ones. Since radioisotope sources are small, a compact, high-efficiency geometry is used so that, despite a low photon output, sufficient characteristic X rays are excited to obtain good counting statistics in about 1 min.

When radioisotopes are used for X-ray fluorescence analysis, the sensitivity at low concentrations is limited by background pulses from Compton and Rayleigh scattering. The background is decreased if the gamma radiation used for excitation has wavelengths that produce, in the region of the detector, scattered radiation of energy different from that being measured. For example, in the analysis of common metals, excitation by 22-kev X rays from ^{109}Cd gives a better signal-to-noise ratio than that by the continuous spectrum from a ^3H–Zr source.

SOURCES AND DETECTORS

Rhodes (paper 13) summarized excitation sources and detectors that are suitable for the determination of various elements (Table II-3).

Table II-3 SUITABLE DETECTORS AND RADIOISOTOPE X-RAY SOURCES FOR ANALYSIS BY X-RAY FLUORESCENCE*

Elements to be determined	Sources	Detectors
K X rays		
Mg† to Si	^3H–Zr (2.0-kev Zr L)	Flow proportional counter
P to V	^{55}Fe (5.9-kev Mn K)	Flow or sealed proportional counter
Cr to Co	^3H–Zr (bremsstrahlung, 5 to 12 kev)	Sealed proportional counter, scintillation counter, ionization chamber‡
Fe to Tc	^{109}Cd (22-kev Ag K)	As above, but proportional counter not used above 20 kev because of low detection efficiency
Co to U§	^{147}Pm–Al (bremsstrahlung, 10 to 100 kev)	
Co to Tm§	^{241}Am (60-kev γ)	
L X rays¶		
Ag to Tm Nd to U	^3H–Zr ^{109}Cd, ^{241}Am, ^{147}Pm–Al	As above for equivalent K X-ray energy

*From paper 13.

†Tanemura[20] reports exciting F and Na K X rays with this apparatus.

‡Ionization chambers are used to detect Fe K X rays (6.4 kev) for "on-line" tin-plate gaging.[21]

§K X rays of energies greater than 50 kev not usually excited because of high scattered background.

¶Mainly by analogy with results for K X-ray excitation.

Three source geometries are described by Cameron and Florkowski (paper 12):

1. Compact geometry (part *a* of Fig. II-2): The source is in the center of the detector window, and the sample is at the optimum distance from the source.[8] A shield keeps direct source radiation from being counted. This arrangement is the most common and gives the highest efficiency.

2. Double-backscatter geometry (part *b* Fig. II-2): In applications where the scattered radiation from the source must be minimum, the source is arranged to excite characteristic ra-

diation of the required wavelength in a suitable target. The source and target are placed in the center of the detector window, as in the first arrangement.[10,18d]

3. Noncompact geometry (part *c* of Fig. II-2): Collimators are used on source and detector, which are some distance apart, and a shield is between the two.[12] The sample is at the intersection of the collimator axes. This system is used only if the higher energy components from the source cannot be prevented from reaching the counter directly when a compact geometry is used (i.e., with sources such as $^{90}Sr-^{90}Y$ bremsstrahlung or with ^{170}Tm).

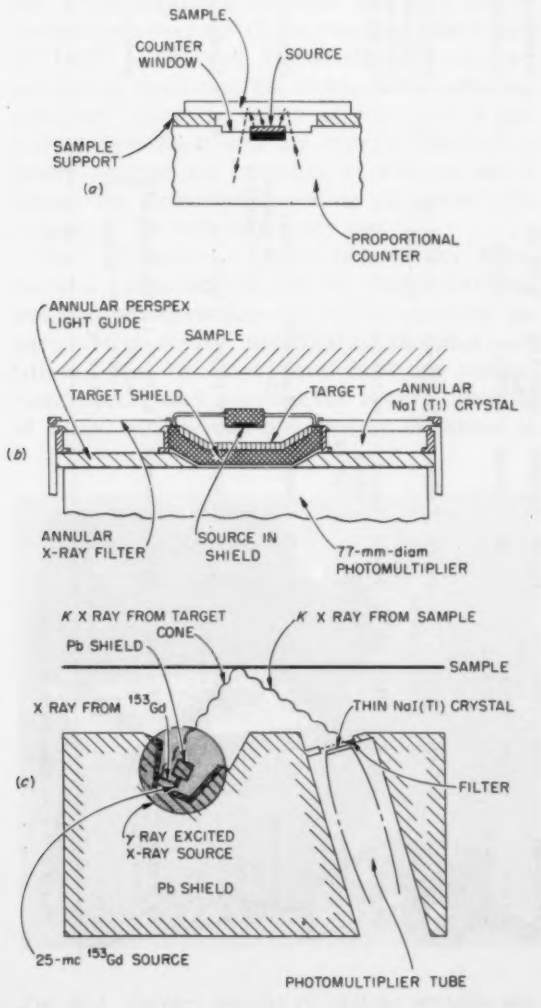

Fig. II-2 Source, sample, and counter arrangements for X-ray fluorescence analysis: (a) compact; (b) double backscatter; (c) noncompact.

APPLICATIONS OF X-RAY FLUORESCENCE

Cameron and Florkowski (paper 12) summarized many of the routine and near-routine uses that have been made of X-ray fluorescence for analysis of various materials (Table II-4). Details of many of these methods are given in the papers presented at the conference and at the Salzburg Symposium on Radiochemical Methods of Analysis.[18]

The determination of silver in photographic film is a particularly useful application of fluorescence analysis. Cameron and Florkowski (paper 12) reported that silver could be determined in unexposed film with a 100-mc ^{147}Pm–Al source without producing a measurable fog on the film. Ezop and Stinchcomb (paper 2) described the determination of silver in radiation-dosimeter films by measuring the silver K peak excited by a ^{147}Pm source. The silver in dosimeter films exposed to 40-kev X-ray doses from 0.001 to 125 r and to ^{60}Co gamma-ray doses from 0.01 to 10,300 r was measured by this technique, and meaningful curves of silver content vs. exposure were obtained. The slopes of the curves at high doses suggest that the method may be used to measure even higher exposures than those investigated. A range of six decades is covered, whereas only two decades can be covered by optical densitometry. With the exception of tungsten in steel, all determinations listed in Table II-4 are for medium-Z elements. Martinelli and Blanquet (paper 8) described methods for determination of heavy metals, e.g., lead in lead ores (galena and cerussite), tungsten in steel, and lead, bismuth, mercury, and tungsten in solutions. In all these measurements they used ^{192}Ir as an excitation source.

For low-Z elements, Martin, Blake, and Morgan (paper 10) gave a preliminary report on the determination of elements with as low an atomic number as fluorine (Z = 9). They used a 3- to 5-curie $^{3}H-Ti$ source and a gas proportional counter with a thin beryllium window. Measurements on elements having K X rays below 1.5 kev (Z < 14) were made in a helium atmosphere to minimize absorption of the soft X rays by air. Ocean-core samples were analyzed for silicon and magnesium. The preliminary results indicated that the effects of other elements in the material being analyzed were even more important in these determinations than with materials of higher atomic weight.

Table II-4 EXAMPLES OF RADIOISOTOPE X-RAY FLUORESCENCE ANALYSIS*

Material	Excited X ray	Source	Filter	Geometry†	Analysis: Range	Analysis: Accuracy	Time, min	Detector	Remarks
Metals and alloys									
Cu–Sn alloy	Cu K	^{3}H–Zr, 2–3 curies		Compact	0.5–99% Cu	0.3% Cu	2	Proportional counter	
Sn–Pb (solder)	Sn K	^{147}Pm–Al, 100 mc		Compact	1–5% Sn	0.25% Sn	1	Proportional counter	
Cu–Zn (brass)	Cu K	^{147}Pm–Al, 100 mc	Ni	Compact	1–99% Cu	0.9% Cu	1	Proportional counter	
Tungsten steel	W K	^{147}Pm–Al, 100 mc	Al	Compact	5–20% W	1% W	0.33	Scintillation counter	
	W K	(^{90}Sr + ^{90}Y)–Pb, 50 mc		Non-compact	0–20% W	0.025–0.45% W	1–3	Scintillation counter	In routine use from 1964
Ores									
Fe ore	Fe K	^{3}H–Zr, 2.5–3 curies	Fe	Compact	0.5–40% Fe	0.2–0.4% Fe	1	Proportional counter (laboratory)	Being tested in industry; in routine use from 1965
						0.5% Fe	1	G–M counter (field)	
Zn ore (0–10% Fe)	Zn K	^{109}Cd, 0.5 mc	Cu–Ni	Compact	1.5–3.5% Zn	0.1% Zn	5	Proportional counter (on stream)	Dilution to 15% solid
	Zn K	^{3}H–Zr		Compact	0–10% Zn	0.2% Zn	2–3	Proportional counter (two-channel)	Being tested in industry; nomographic interpretation
	Fe K	^{3}H–Zr, 2.5–3 curies		Compact	0–10% Fe	0.2% Fe			
Zn concentrate	Zn K	^{109}Cd, 0.5 mc	Cu–Ni	Compact	40–65% Zn	2% Zn	5	Proportional counter	Dilution to 15% solid
Zn tailings	Zn K	^{109}Cd, 0.5 mc	Cu–Ni	Compact	0.05–0.5% Zn	0.02% Zn		Proportional counter	
Cu ore (0–10% Fe)	Cu K	^{109}Cd, 0.5 mc	Fe–Ni	Compact	0–7% Cu	0.1% Cu	5	Proportional counter	Combined absorption-fluorescence used to overcome matrix effect
	Cu K	^{3}H–Zr, 2.5–3 curies	Co–Ni	Compact	0–15% Cu	0.1–0.2% Cu 0.3–0.5% Cu	5 3	Proportional counter (laboratory) G–M counter (field)	In preparation for routine use in 1966; nomographic interpretation
Sn ore	Sn K	^{241}Am–Cs, 4 mc	Pd–Ag	Double back-scatter	0–10% Sn			Scintillation counter	Combined absorption-fluorescence used to overcome matrix effects
Other materials									
Co in hydrocarbons (up to 0.13 g of Fe per liter)	Co K	^{147}Pm–Al, 100 mc ^{3}H–Zr, 2–3 curies	Mn–Fe	Compact	10 ppm–2% Co	1% relative (10% relative for 50 ppm Co)	2	Proportional counter	
Fe in oils	Fe K	^{3}H–Zr, 2–3 curies	Fe	Compact	0–1% Fe	0.03% Fe	1	G–M counter	Being checked for routine use; mixing to avoid sedimentation of Fe suspension
Ag in unexposed film	Ag K	^{147}Pm–Al, 100 mc	Rh–Mo	Compact	0.077–38.4 mg/cm^2 Ag 1–50 g/sq ft Ag	0.015 mg/cm^2 0.2 g/sq ft	1	Scintillation counter	Under permissible fog level

*From paper 12.
†See Fig. II-2 for description of geometry.

PORTABLE X-RAY FLUORESCENCE ANALYSIS INSTRUMENTS

An interesting feature of the papers on X-ray fluorescence analysis was the description of two portable instruments for field use.

Rhodes (paper 13) discussed a portable analyzer, consisting of a scintillation counter and a transistorized battery- or line-operated rate meter, which was developed in conjunction with Hilger and Watts Ltd. and was to be commercially available early in 1965. Figure II-3 shows the instrument proposed for factory and laboratory use; a tropicalized "geological" version is also being manufactured for field use. The main features of the rate meter are backing-off and scale-expansion controls that give nearly continuous coverage of the counting-rate range 10^2 to 10^5 per second. The scintillation-counter measuring head uses the double-backscattering geometry shown in part *b* of Fig. II-2. It has interchangeable filters for energy selection, a safety shutter for radiation protection, and a means for accurately locating the probe with respect to the material under test.

The instrument can be adapted for beta-particle backscattering, X-ray backscattering, and X-ray fluorescence by suitably choosing the radioisotope source, scintillation phosphor, and filters. The "factory" version of this instrument should find wide use for routine checking of alloy composition and coating thickness in the metallurgical, plastics, and electronics industries. With the geological version, which has a probe designed to accommodate measurement of rock with surface roughness of ±0.2 in., ore strata can be mapped in situ. The same instrument is intended for field assay of core, rock, and powdered-ore samples.

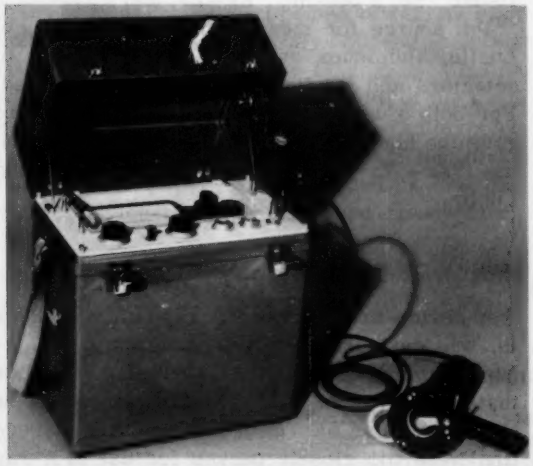

Fig. II-3 Factory version of isotope portable analyzer showing the standard probe for measurements on flat surfaces. (Courtesy of Hilger and Watts Ltd., Camden Town, London, England. From paper 13.)

Karttunen and Henderson (paper 11) described a portable X-ray fluorescence apparatus (Fig. II-4) of recent design. This instrument operates

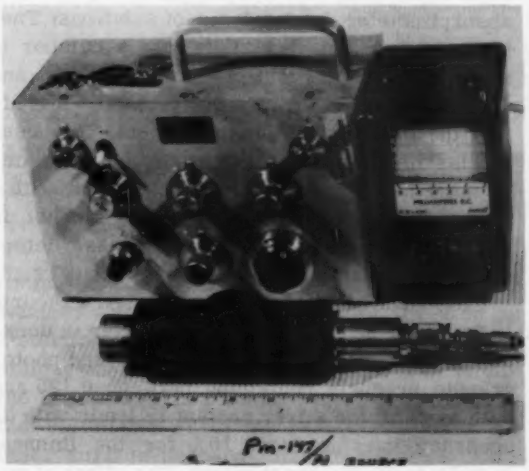

Fig. II-4 Portable fluorescence X-ray apparatus that occupies approximately 0.5 cu ft volume and weighs 11 lb. (From paper 11.)

with either a $^{3}H - Zr$ or a $^{147}Pm - Al$ source. A proportional-counter tube with a 2-mil beryllium window and 90% Kr–10% CH_4 counting gas is used, and miniaturized and transistorized circuits are supplied with power from a nickel–cadmium battery with a built-in charger. Data are plotted on a small recorder. This apparatus has been used successfully to analyze a number of standard quaternary mixtures of varying amounts of CaO, V_2O_5, Fe_2O_3, and CuO, synthetic and NBS portland cements, and various alloys.

X-RAY ABSORPTIOMETRY

The absorption of X rays as a method of chemical analysis has received more attention in the past than X-ray fluorescence,[19] and sulfur is routinely determined in hydrocarbons by X-ray absorption using ^{55}Fe and $^{147}Pm - Al$ sources.[22,23]

Only a few papers at this conference discussed X-ray absorption as an analytical method. Cameron and Florkowski (paper 12) mentioned that, in the analysis of copper and tin ores, a combination of X-ray fluorescence and absorption was used to reduce inaccuracies in the copper and tin determinations caused by other elements in the ores (Table II-4). These workers also determined lead in lead ores by using a $^{90}Sr-^{90}Y$ source and a lead target.

Preuss, Collins, and Kann (paper 4) have been developing and evaluating a simple X-ray absorptiometer for analysis of solutions. They have made a detailed study with a number of sources, including $^{147}Pm-Mo$, $^{147}Pm-Sn$, and $^{147}Pm-Cu$, to determine the variables that affect the operation of the instrument. They used the instrument with a $^{147}Pm-Sn$ source to determine tetraethyl lead in gasoline and with a $^{147}Pm-Sn$ source to determine alcohol in alcohol–water mixtures, making these determinations with a probable error of about 1%.

Ezop and Stinchcomb (paper 2) used a 1-curie $^{147}Pm-Sb$ source to determine silver in unexposed photographic film. They analyzed photographic products containing between 0.125 and 2.25 mg/cm^2 for silver content in 1 min with an accuracy varying from 10% for the thinnest sample to 1% for the thickest. The effect of radiation dose on the fog level of the film was within limits considered tolerable, provided the increase in optical density was uniform.

Industrial Gaging

Hitch (paper 1), in his introduction to the Symposium, pointed out that the program to develop low-energy gamma emitters was stimulated mainly by the needs of the gaging industry in the early 1950's. The few papers presented on industrial gaging were, in general, parts of broader survey papers.

Badgett (paper 5) discussed some of the basic considerations that go into the design of an industrial radiation-absorption measurement gage. He reviewed the requirements for radiation sources in these instruments and showed how much more easily isotopic sources meet the requirements than does an X-ray machine. He also discussed the design of a gage for measuring liquid hydrogen flowing in a pipe and showed that the sensitivity of measurement is directly related to the source energy. His paper indicates that the trend in industrial radiation gages will be the obtaining of greater specificity by absorption of monochromatic gamma or X rays or by measurement of X-ray fluorescence in a process stream.

Cameron and Florkowski (paper 12) discussed the continuous determination of coating thickness by X-ray fluorescence. They pointed out that coating thickness can be determined by measuring either the increase in intensity of X rays excited in the coating as the thickness increases or the decrease in intensity of X rays excited in the substrate as it is attenuated by a coating of increasing thickness. They gave a number of examples of the measurement of coating thickness by X-ray fluorescence (Table II-5) and stated that almost all electroplated tin produced in the United Kingdom is now controlled by radioisotope gages.

An interesting gaging application described was the measurement of the uniformity of wall thickness of precision stainless steel and Zircaloy tubing (Locher, paper 9). Tubing of this type sometimes shows excessive variation in wall thickness because the inner and outer walls, although both perfectly round, are not concentric. A ^{241}Am source was inserted into the tube, and the radiation was measured outside. By setting up special jigs that provided for free rotation and translation of the tube, the entire tube wall could be measured to within ±0.0001 in. This type of gage has now been in successful operation for over 4 years.

Ezop and Stinchcomb (paper 2) evaluated a 10-mc ^{90}Sr, a 500-mc $^{85}Kr-Zr$, and a 250-curie ^{147}Pm source for measuring the thicknesses of Lucite, aluminum, and steel. Several types of detectors and reading instruments were used. For both aluminum and steel, the ^{147}Pm source gave the highest sensitivity and readability, but the $^{85}Kr-Zr$ source was best for measuring Lucite.

Industrial Radiography

A device that provided a resolution of 0.002 in. was described for 360° panoramic radiography of critical joints in special steel tubes that ranged from 0.25 to 1 in. in diameter (Locher, paper 9). The tubes requiring inspection were too closely packed for use of X-ray equipment or sensors of other conventional nondestructive testing devices.

High-resolution radiography at source-to-film distances of only $\frac{1}{8}$ to $\frac{1}{2}$ in. has been im-

Table II-5 COATING-THICKNESS MEASUREMENT BY X-RAY FLUORESCENCE*

Material	X ray	Source	Filter	Thickness range, cm	Accuracy
Electroplated Sn on steel	Fe K, 6.4 kev	^{3}H–Zr		$5-180\times10^{-6}$	1% in 10 sec
Electroplated Zn	Fe K, 6.4 kev	^{3}H–Zr	Ni, 0.0013 cm	$1.3-30\times10^{-5}$	2% in 10 sec
Hot-dipped Zn on steel	Zn K	^{241}Am	Ni + Co, both 0.0013 cm	$1.8-4.5\times10^{-3}$	2% in 100 sec
Pt or Au on Ti	Ti K, 4.5 kev	^{3}H–Zr		$5-130\times10^{-6}$	2% in 40 sec
Ni on Cu	Ni K, 7.5 kev	^{3}H–Zr	Co, 0.0013 cm	$1.5-15\times10^{-4}$	2% in 100 sec
Cu on steel	Fe K	^{3}H–Zr	Fe, 0.0013 cm	$0.8-13\times10^{-4}$	2% in 200 sec
Ag-plated Cu wire	Cu K, 8.0 kev	^{3}H–Zr		$10^{-6}-10^{-4}$	10% in 10 sec
Ink on paper	Fe K	^{3}H–Zr		$0.4-4\times10^{-5}$	5% in 30 sec
Ink on Al printing plate	Fe K	^{3}H–Zr		$0.4-4\times10^{-5}$	5% in 30 sec
Glaze coating on paper	Ti K	^{3}H–Zr		$2.5-50\times10^{-5}$	5% in 30 sec
Pressure-sensitive coating on paper	Ca K, 3.0 kev	^{3}H–Ti		$2.5-50\times10^{-5}$	2% in 10 sec
Alkaline-earth carbonates + Ni on Ni	Sr K, 14.2 kev	^{147}Pm–Al	Al, 0.015 cm	$3.5-13\times10^{-4}$	5% in 10 sec
Ag-coated electrical contacts	Ag K, 22 kev	^{147}Pm–Al	Al, 0.04 cm	$0.5-5\times10^{-3}$	5% in 40 sec

*From paper 12.

possible except with microscopic sources and extremely long exposure times. Coleman et al. (paper 14) showed that the relatively large focal spot in the usual radioisotope source was a major drawback to high-quality radiography, and radiographs from a point source would be greatly distorted because of oblique incidence of the rays. This problem was solved with a special exposure device (Fig. II-5). A long, thin ^{241}Am gamma source was centered in a series of "washers," alternately lead (or depleted uranium) foil and paper. The assembly puts out many very narrow radial beams of gamma rays and is equivalent to narrowly collimated beams from many X-ray tubes in tandem used inside the tube being measured. The width of the focal spot is the inside diameter of the radioisotope capsule, and the length of the spot is the thickness of the paper spacer. When in use, the entire device is either advanced slowly along its major axis or is slowly oscillated to remove shadows of the collimator disks. There is no distortion on the radiographs resulting from oblique incidence of the radiation beam. Within the angular-spread limits set on each beam by the collimator slits, the radiation strikes the inner surface perpendicular to the axis of the tube being examined, as if the source were at infinite distance.

Resolution, i.e., the ability to detect small defects, is related to the geometrical unsharpness of the radiograph and is controlled by the design of the device. As the spacer thickness was decreased, unsharpness decreased, but the gamma dose rate, which could not be measured because of the strange collimation, also decreased.

The ability to detect small variations in absorption by the specimen is closely related to the energy spectrum of the radiation used and

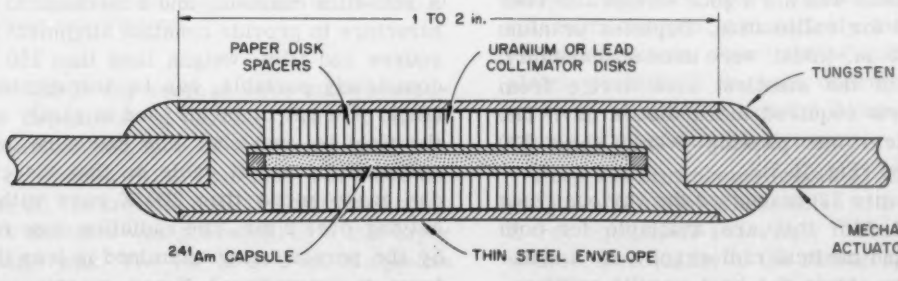

Fig. II-5 Panoramic exposure device. (From paper 9.)

the absorption characteristics of the metal under inspection; the photon energy needed was about 60 kev. A bremsstrahlung source and a ^{75}Se source were tested, and both were unsatisfactory because of the hopelessly low output of the low-activity bremsstrahlung source and the energy spectrum of the ^{75}Se. Fortuitously, the U. S. Atomic Energy Commission (AEC) released ^{241}Am under Byproduct Material Licenses and saved the project from impracticability, if not failure. Americium-241 gamma rays are essentially monochromatic, about 60 kev, and equivalent to the spectrum of a 75-kvp X-ray machine; they are ideal radiation sources. Both the energy spectrum and the 458-year half-life are highly satisfactory because fabrication of these devices takes a long time, and source replacement every few months is difficult and expensive.

Although the output of thin ^{241}Am gamma sources in a typical capsule at 1 m is only 6 to 8 mr/(hr) (curie) and the linear self-absorption coefficient of ^{241}Am is probably greater than 60 cm^{-1} for 60-kev photons in solid americium, the use of long exposure times with high-contrast films was of far less concern than inability to do the job at all. With source-film distances of 1 cm or less, exposure times with 200 mc of ^{241}Am in a capsule of 1.5 in. active length were 6 to 10 hr with Eastman M film for 0.1-in. steel, but a resolution of 0.0015 in. was readily obtained. The clearance between the outside of the source device and the inside of the tube being radiographed should be as small as possible. Teflon slide bearings slightly larger than the outside diameter of the device were used as spacers and guides to prevent metal-to-metal contact with the tube. A small scratch in these tubes might necessitate their replacement.

With very small devices, where the effective length of the collimators was as small as 0.045 in., lead was not a good enough absorber to be used for collimation. Depleted uranium disks, 0.005 in. thick, were used successfully. Assembly of the smallest size device from finished parts required more than a day. The main problem was packing evenly some 200 washer pairs (Fig. II-5).

Frede (paper 22) discussed the various types of Polaroid film that are available for both industrial and medical radiography. He emphasized that to obtain the best results a radiographer must understand the sensitivity of the various films both to X and gamma rays and to visible light. The response to visible light is particularly important when a fluorescent intensifying screen is used. He also reported some preliminary experimental work in which X-ray diffraction patterns were recorded on Polaroid film. With special Polaroid materials, usable diffraction patterns were obtained in 30 min. Normal wet-process film-exposure time under the same conditions would be 9 to 10 hr.

Ezop and Stinchcomb (paper 2) showed a series of radiographs of various items of electronic equipment taken with a ^{147}Pm source and both with Kodak X-ray film with an intensifying screen and with Polaroid film. Exposures of 20 sec to 5 min were required, and source-to-film distances were 6 to 12 in.

Medical Uses

Three types of medical applications of low-energy sources were presented: (1) for medical radiography (papers 2, 16, 21), (2) for gaging to measure mineral content of bone and fat content of the body (papers 17–19), and (3) for diagnosis by scintillation scanning (papers 15, 20).

MEDICAL RADIOGRAPHY

Green (paper 21) described his work on the development of portable radiographic devices containing ^{169}Yb sources. This is an outgrowth of the work of Krabbenhoft and Green[24] evaluating a number of isotopic sources for radiography. They compared the quality of the radiographs obtained with those from the usual diagnostic X-ray machine and concluded that ^{169}Yb was the best source for medical radiography available at the time.

Green and Krabbenhoft[25] and Allen et al.[26] have described a portable system for making field chest X rays. This entire system, which includes a 75-curie ^{169}Yb source, a source holder, a roll-film cassette, and a mechanical support structure to provide constant alignment between source and film, weighs less than 250 lb. It is completely portable, can be transported in the trunk of a car or by air, and is easily set up in the field by one man. The exposure device is operated by a 6-ft cable so that the operator can make more than 200 X rays without receiving over 2 mr. The radiation dose received by the person being examined is less than that from a conventional X-ray machine. One of these units was recently used by workers of the

U. S. Agency for International Development for a chest survey of an Indian tribe in the upper Amazon region of South America, where there is a serious tuberculosis problem.

Although the half-life of ^{169}Yb is only 32 days, a 75-curie source (original strength) can be used for approximately 3 months by increasing exposures to compensate for source decay. Spent source-container units are returned to the factory for replacement so that sources do not have to be handled in the field.

The source container unit can also be used for other diagnostic work in the field with a conventional cut-film cassette. Both the source unit and the cassette are mounted on tripods, and, of course, facilities are needed for developing the film. (With the roll-film cassette used for chest X rays, the film can be processed at a remote location sometime after the exposure.) Radiographs have been made of the chest, hand, elbow, leg, pelvis, and other parts of the body with such units.

Söremark (paper 16) described a unit with a ^{125}I source for intraoral radiography. He outlined the requirements that a radioisotope source for radiography should have:

1. The specific activity should be as high as possible, i.e., the carrier-free isotope should be available. This is the case with ^{125}I.
2. The intrinsic yield of usable radiation should be as high as possible. Being considerably more than 100%, the X-ray yield from ^{125}I is superior.
3. The half-life should be short because it is inversely proportional to the activity/weight ratio in the case of carrier-free activities, but, for obvious practical reasons, it should not be too short. The 60-day half-life of ^{125}I seems to offer a well-balanced compromise.
4. The source should be composed of as few and as light elements as possible in addition to the radioactive one. The elemental form would thus be the best choice, but in practice choice is limited by safety considerations to stable compounds of very low vapor pressure. The compound should also allow easy fabrication into a small-sized source. In the case of ^{125}I, silver iodide and copper iodide seem to fulfil these requirements.

Iodine-125 X-ray sources were made by electrolytic deposition of carrier-free ^{125}I on the hemispherical end face of a 0.5-mm-diameter silver or copper wire. This face was made the anode in an electrolytic cell. Deposition on other parts of the wire was prevented by a thin layer of paraffin wax. The $Na^{125}I$ in 2 ml of aqueous sodium hydrogen sulfite solution was efficiently stirred during the deposition.

Copper is a better backing material than silver for two reasons: self-absorption of the X rays is less in copper iodide than in silver iodide, and the secondary radiation produced in copper is of lower intensity and has a lower energy than that from silver. Therefore the secondary radiation of copper is more easily attenuated to a negligible level.

After electrolysis the active tip of the wire was covered with an air-hardening epoxy resin to prevent mechanical loss of the ^{125}I. The wire was then mounted in a cavity of a brass rod, which was inserted in a gold cylinder that served as shielding material. The aperture in the gold cylinder was covered with plastic and an aluminum foil to absorb electrons and soft X rays. The brass rod may be moved back and forth in the gold shielding. The radiation source is moved into exposure position by a photographic shutter-release cable and is brought back into nonexposure position by a spring.

Iodine-125 can be concentrated to a very small source. Thus the distance between source and object may be short, and exposure times are reduced without impairing the quality of the roentgenograms. Various types of exchangeable collimators are used to diminish unsuitable radiation. Ordinary intraoral roentgenograms can easily be made with a simple film–source holder attached to the teeth. The source unit is also small enough to make panoramic roentgenograms by placing it intraorally in a holder and collimator—likewise fixed to the teeth. The collimator is so constructed that radiation is allowed forward and laterally, giving an exposure of one or both jaws on the extraorally placed film.

Exposure data for various human objects are listed in Table II-6. As opposed to conventional X-ray apparatus, this "pocket X-ray unit" requires minimum space and is light, easy to transport, rugged, and entirely independent of electrical power. For it to be used as a completely portable X-ray unit, darkroom facilities must be eliminated, and the Polaroid technique seems to meet this requirement.

Ezop and Stinchcomb (paper 2) showed radiographs of a skeleton hand taken with conventional X-ray equipment and with a ^{147}Pm beta-excited source. The quality of the radiograph taken with the ^{147}Pm source was not so good as that taken with the conventional equipment.

MEDICAL GAGING

The mineral content and density of bone have been determined for a number of years with conventional X-ray machines and film by measuring the density of the resulting radiograph.[27,28] Mayer et al.[29] improved this technique by measuring the intensity of the transmitted rays from an X-ray machine with a scintillation counter and thus eliminated the errors introduced by standard film development and densitometry. However, this procedure still left many uncertainties.

Table II-6 EXAMPLES OF EXPOSURE DATA FOR SOME NO-SCREEN FILMS*

Object	Film	Source-to-film distance, cm	Exposure times for a 1-curie ^{125}I source, sec
Panoramic roentgenogram of lower and upper jaw	Kodirex	6	4
Three teeth	Kodak ultraspeed dental film	7	8
Hand	Cea Singul-X	20	20

*From paper 16.

Cameron and Sorenson[30] suggested using radioisotope sources in this technique for measurement of composition of body materials, and the three papers on this subject at this symposium were based on their work. They suggested that the use of isotopes results in the following advantages over the older techniques:

1. The transmission of the photon beam is measured directly by counting techniques, using a scintillation detector system.
2. The photon beam is essentially monochromatic.
3. The photon beam and detector are well collimated.
4. The effects of the tissue around the bone are taken into account.

Lanzl and Strandjord (paper 17) presented a method for in vivo determination of the mineral content of bones in humans and small animals. In the work with humans, they measured X-ray transmission of the second phalanx of the third digit of the left hand since the overlying soft tissue is at a minimum and the finger is easy to immobilize. With small animals, the rear leg bone was measured.

The finger to be measured was held immobile by two plastic plates, which were milled out in the region of measurement but quite thick elsewhere. The ^{125}I source was prepared by adsorption of the iodine from solution on an ion-exchange resin. This resin was then placed in a plastic cylinder, which in turn was put in a brass source shield with a sliding shutter. The beam was collimated by a brass insert, and beam diameters from 0.3 to 2.0 mm were used.

The beam was detected by a 2-mm NaI(Tl) crystal. To prevent light and moisture from entering the crystal, it was covered on the entrance side by a 0.13-mm beryllium foil, which absorbed only about 1% of the radiation. Since the photon energies from ^{125}I are low, the crystal was a very efficient detector from which very little energy escaped. To reduce the background the crystal was surrounded by another shield, which had a 3-mm-diameter collimator hole.

A conventional radiograph of the finger held in a slotted device marked in the same way as the locator on the measurement device (Fig. II-6) permits accurate location of the desired point for measurement.

The unit was normally programmed to make transmission measurements across the entire width of the finger automatically. The finger was held immobile, but the source and detector were mounted on a frame that was driven laterally across the finger by a motor. Measurements were made at 1-mm intervals. During each measurement the source and detector were held fixed. During the time the source and detector were moving, the digital recorder printed out the time or the counts for the previous interval. Upon completion of the 1-mm movement, the measurement was repeated. This process was continued until the entire width of the finger had been scanned.

The device is being used to study osteoporosis in postmenopausal females and other conditions

that result in changes in the mineral content of the bone. Preliminary results indicate that the device will be valuable in such studies.

Goodman and Levin (paper 19) also measured the mineral content of bone by measuring the absorption by bone of the fluorescence X rays produced by a ^{147}Pm source from barium (32 kev) and from platinum (67 kev). The low-energy rays are absorbed much more strongly by bone than by an equivalent amount of soft tissue, but the high-energy rays have mass absorption coefficients that are not nearly so different for the two tissues. The procedure was described only briefly.

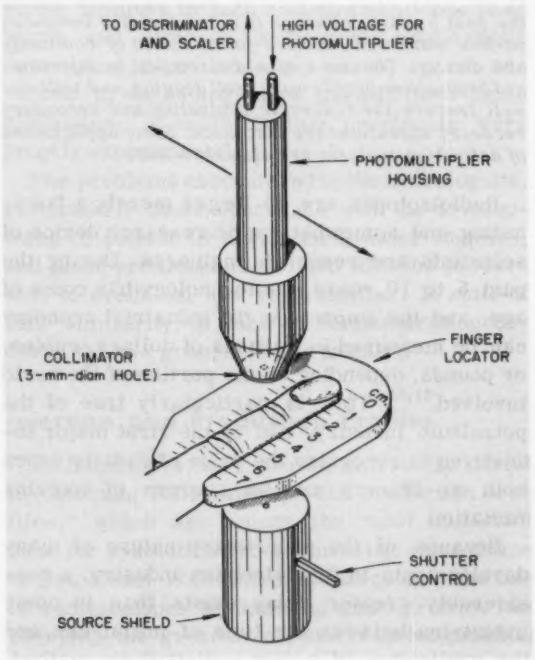

Fig. II-6 Diagram of equipment for studying bone mineral content in vivo.

Sorenson and Cameron (paper 18) reported the use of X-ray attenuation at two energies to measure the amount of fat in tissue. They showed that

$$\frac{(\ln I_0/I) \text{ at } 27.4 \text{ kev}}{(\ln I_0/I) \text{ at } 59.6 \text{ kev}} = kf_{fat}$$

where I_0 and I are the incident and transmitted intensities of the radiation at each energy, f_{fat} is the weight percentage of fat in the sample, and k is a proportionality constant. The 27.4-kev gamma was produced by ^{125}I and filtered through tin, and the 59.6-kev gamma was produced by ^{241}Am. The method was tested on mixtures of sodium acetate and polyethylene, which simulate muscle and fat, and on samples of animal muscle and fat. The results were within 3% of other independent determinations, and the technique was judged to be accurate for determining the composition of such mixtures.

ORGAN VISUALIZATION

Wagner (paper 15), in his introduction to the medical section of the Symposium, and Ter-Pogossian (paper 20) reported the use of isotopes for visualization of organs or structures for diagnostic purposes. Ter-Pogossian lists the following advantages for low-energy X or gamma rays for organ visualization:

1. Particularly efficient collimators may be used.
2. The photons are scattered nearly isotropically, while higher energy photons tend to be scattered in the forward direction. This isotropic scattering, associated with the absorbability of lower energy photons, reduces the intensity of the scattered radiation that reaches the detector and thus decreases a particularly undesirable component of the noise introduced in the examination.
3. In most cases, the low-energy photons dissipate less energy in the patient than do higher energy photons.
4. Several of the isotopes that emit low-energy X or gamma rays, and have already found many applications in nuclear medicine, do not emit beta rays, and they deliver a lower radiation dose to the patient than do those that emit high-energy radiation accompanied by beta rays.
5. Because of the high absorbability of low-energy photons in matter, detectors with relatively low cross-sectional density may be used. This property allows the design of the scintillation camera discussed here.

Both Ter-Pogossian and Wagner used an X-ray image amplifier rather than the usual scintiscanner. An X-ray image amplifier is a device that converts the energy of X- or gamma-ray photons to light and multiplies the light photons by a large factor, typically about 6000. The input screen of conventional X-ray amplifiers does not offer much stopping power for high-energy photons, such as those emitted by most radioactive isotopes; therefore the efficiency of the instrument for detecting such radiation is low. On the other hand, this instrument was designed, and is very suitable, for detection of photons with energies less than about 150 kev.

The system used is shown in Fig. II-7. The image amplifiers had an 8-in.-diameter input and a gain of about 2400. The output screen in all cases was 1 in. in diameter. The image provided by the X-ray amplifier is projected onto a Polaroid camera of conventional design by a fast-lens system with a numerical aperture slightly less than 1. Polaroid 410 film with an American Standards Association rating of 10,000 was used.

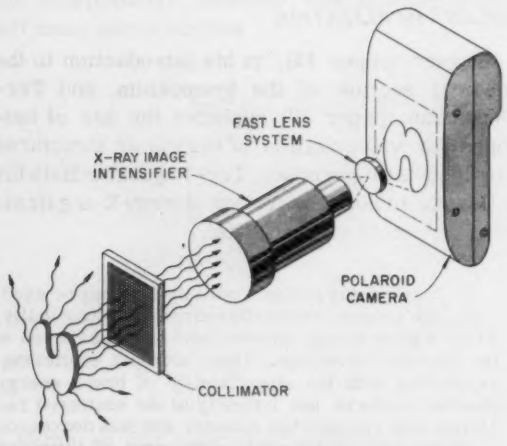

Fig. II-7 Autofluorographic system for visualization of the thyroid. (From paper 20.)

Ter-Pogossian tested the system with different phantoms simulating organs most commonly visualized by scintiscanning: (1) a thyroid phantom with suitable defects containing ^{125}I, with which satisfactory images were obtained in approximately 3 min when the ^{125}I content was about 0.5 $\mu c/cm^3$; (2) a phantom simulating a head containing a tumor with ^{197}Hg, with which suitable images were obtained in 15 min when the ^{197}Hg content was about 2 $\mu c/cm^3$; and (3) different containers simulating lungs containing ^{133}Xe. Tests were also made on patients to visualize kidneys with ^{197}Hg. The results indicate that pictures of diagnostic quality can be obtained in shorter times than are required with conventional scintiscanning systems.

Wagner used an early model of the image-amplifier system to make some preliminary studies of the localization of ^{99m}Tc and did extensive studies with ^{133}Xe on measuring the flow of blood to various parts of the body.

(Robert H. Lafferty, Jr.)

Recent Advances in Radioisotope Applications for Exploitation of Petroleum and Natural-Gas Reservoirs*

By F. E. Armstrong and W. D. Howell†

This paper reports some of the more important recent advances in the application of radioisotopes to the exploitation of petroleum and natural-gas reservoirs. No attempt was made to summarize all work in the field. Most references were published during the past 5 years, although in a few isolated instances earlier work was included for the sake of continuity and clarity. The uses of radioisotopes in injection- and production-profile work, well logging, and well-to-well tracers for reservoir evaluation and secondary recovery operations are discussed. Some applications of activation analysis are also reviewed.

Radioisotopes are no longer merely a fascinating and somewhat exotic research device of scientists and research engineers. During the past 5 to 10 years, the technology has come of age, and the impact on the industrial economy can be measured in millions of dollars, rubles, or pounds, depending on the portion of the world involved.[31,32] This is particularly true of the petroleum industry, one of the first major industries to recognize the value of radioisotopes both as tracers and as sources of ionizing radiation.

Because of the proprietary nature of many developments in the petroleum industry, a considerably greater delay exists than in other industries between the time of initial use and the publication of a new radioisotopic method. This is especially true when compared to biological science and hydrology, for example. Even so, much literature on the subject has accumulated during the past few years, particularly in the United States and the Soviet Union.[33]

Radioisotopes are used in various ways to obtain information useful to the engineer in developing productive reservoirs of oil or natural

*Papers presented by the U. S. delegation at the Third Symposium on the Development of Petroleum Resources of Asia and the Far East, Tokyo, Japan, November 1965.

†Bartlesville Petroleum Research Center, Bureau of Mines, U. S. Department of the Interior, Bartlesville, Okla.

gas and in drilling and completing wells. First, as tracers they permit the engineer to follow the movement of liquids or gases within a wellbore or through the reservoir. By adding tracers to various materials that are emplaced in the wellbore, he can determine their ultimate location. Second, the use of gamma sources permits measuring the density of gaseous, liquid, and solid materials within or surrounding the wellbore. Third, the Be(α,n) reaction produces neutrons that may be used in many ways, and bombarding a tritium target with deuterons produces an even more intense source of high-energy neutrons. Although the basic knowledge required for application of many of the principles involved in such measurements has been known and some of the reported applications have been in use for many years, recent advances in technology now permit the routine application of methods that previously were largely experimental.

The problems encountered by the hydrologists, particularly those concerned with the development of potable or agricultural water supplies, and those problems associated with the production of crude oil are very similar. In spite of this similarity, a lack of communication between the two groups seems to exist.

Injection and Production Profiles

Radioisotopes have proved extremely useful in obtaining fluid-injection and production profiles,* which are among the most important data for evaluation and engineering exploitation of petroleum reservoirs. Consequently the use of radioisotopes in obtaining these profiles has received the greatest amount of developmental effort.

Three basic methods for determining fluid-injection or production profiles have evolved over the past 10 to 15 years: (1) plating or adsorption methods in which radioactive indicators are plated out or adsorbed in vertical zones of maximum (or in some instances, minimum) fluid injection or production; (2) measurement of fluid-flow ratios between different vertical sections of the wellbore (the interface method); and (3) direct measurement of fluid flow within the wellbore.

Radioisotopes have been used in various ways in all these methods. A fourth method, although not directly quantitative, determines the points of entry of fluids by measuring the fluid density with a gamma-ray device,[34] by measuring the neutron absorption of the fluid,[35] or by determining a constituent of the fluid by neutron activation.[36]

PLATING METHOD

One of the earliest techniques for determining liquid-injection profiles, particularly in water-injection wells, is the so-called "plating technique."[37,38] A gamma-emitting radioisotope, often ^{46}Sc but sometimes ^{60}Co, ^{192}Ir, ^{131}I, or ^{110}Ag, is incorporated into or onto the surfaces of small particles of a material, usually plastic, whose density is carefully chosen to match that of the injection water. The particles must be small enough to ensure their traveling with the injected liquid but large enough to prevent penetration of the sandface to beyond the detection limit.

The particles are preferably released from a suitable device within the well immediately above the zone of interest, or they may be injected into the flow stream at the ground surface. From this point the particles are carried along by the flowing liquid, and, hopefully, the amount distributed along the sandface can be directly related to the amount of liquid entering the formation. A gamma log made at this time with a conventional gamma-ray well probe is compared with a background log made before the test to indicate the vertical injection profile of the well.

The method is fast and, if carefully applied, yields excellent results. Variations in well diameter have less effect on the results than in methods involving direct flow measurements and may often be neglected, particularly with the use of radioisotopes having high-energy gamma emissions, such as ^{46}Sc or ^{60}Co; this eliminates the requirement for a caliper survey.

There are two primary disadvantages. First, unless the particles are very carefully selected for proper density, the results will indicate that all liquid is leaving at either the top (particles not dense enough) or the bottom (particles too dense) of the test zone, and there is no way of detecting such errors. Second, the well is

*A fluid-injection profile determines the points of fluid entry into the sand face from the wellbore (i.e., the sand sections that are accepting injection fluid). A fluid-production profile determines the points of fluid entry into a wellbore (i.e., the sand sections that are producing formation fluids).

contaminated by the added radioisotope. Although ^{46}Sc (84 days, 0.89- and 1.12-Mev γ's) is a good compromise for a tracer, its half-life is undesirably long, and residual radioactivity may prevent another radioisotope survey for many months. Iodine-131 (8 days, 0.36-Mev γ) does not present as much difficulty in this respect, but the much lower energy of the emitted gamma ray is absorbed more rapidly in the well fluid and thus necessitates considerably more accurate knowledge of variations in the wellbore diameter.

INTERFACE TECHNIQUES

The most accurate, reliable, and widely accepted method of determining profiles for water-injection wells is the interface method described by Pfister[39] in 1948 and since modified. Its application is improved by the use of radioactive tracers. In this method liquid, usually water or brine, is injected through at least two separately metered systems, one at the top and the other at the bottom of the zone of interest. The interface between them can be determined by tagging one or the other, by introducing between the two layers a "slug" of tracer that is miscible with one fluid but not with the other, by using two liquids of different densities such as fresh water and brine, or by some other suitable method. The position of this interface, together with data on the variation of flow rates of the two systems as the interface is moved, permits calculation of the well profile. In a variation of the method, fluid is injected into only the upper (or, more rarely, the lower) zone, and the rate of movement of the interface is recorded.[40]

Limanek[41] reversed the technique for determining production profiles by running two "strings" of tubing into the well (Fig. II-8), establishing three measured flow streams from the well, and positioning the two interfaces thus formed in the wellbore at the top and bottom of the zone of interest by throttling the flow rates from the three streams at the surface.

Wood[42] suggested a technique that permits making several interface tests with radiotracers as interface markers but prevents the residual tracer from interfering with the subsequent test. Tracers with increasingly higher gamma-ray energies are used and identified by spectrometric logging techniques.

McKay[43] suggested the addition of a nuclide that has a large neutron cross section and forms a short-half-life isotope (manganese, for example) to one injected fluid. The added tracer material is then determined in the wellbore by activation with an isotopic neutron source. This eliminates the hazard of handling radiotracers in one of the injected fluid streams but introduces another hazard, that of handling a quite large neutron source.

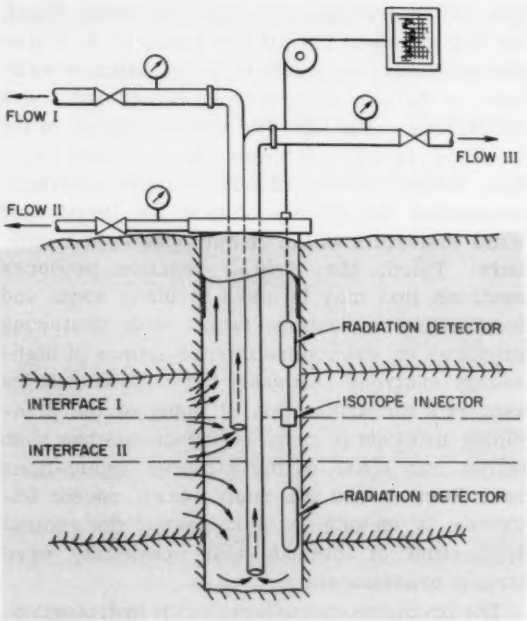

Fig. II-8 Limanek's[41] technique for determining well production profile.

FLOW-RATE METHODS

The measurement of flow rate within the wellbore requires less preparation than does the interface technique and does not require running additional tubing into the well. However, it does require an accurate knowledge of the wellbore cross section; thus a caliper log must be made in addition to the flow tests. Early applications of the method required relatively high flow rates because of the insensitive instruments for making measurements, which were limited at first to spinners, i.e., small propellers turned by the moving fluid. This method is still very useful in wells in which the flow rate is high. In other wells, particularly those in small, marginal fields where the water-injection rate is from tens to a few hundred barrels per day, more sensitive meth-

ods are required, and radioisotope techniques have been most useful.

Basically most of these methods involve injecting a small amount of radioactive material, usually a solution physically similar to the wellbore fluid, and timing the rate of travel of the activity over a measured vertical interval within the wellbore.[44] Occasionally two detectors are used, and the time of travel between them is measured.[45] More commonly, however, a single detector is used, and the time interval is measured from the moment of release of the radioisotope until the detector indicates its passage.

A number of ingenious methods have been devised for release of small, controlled amounts of tracer. One of the earliest of these was a glass ampule, which was broken at the proper time and location by a solenoid device.[46] This technique is simple and foolproof, but only a very few operations can be completed with one trip of the tool into the well. Subsequently, motor- or solenoid-driven positive-displacement pumps were developed which can deliver on command as many as 100 portions of tracer before reloading.[47] Other methods include release of the radioisotope through a porous membrane,[48] "deplating" the isotope from a solid source (^{60}Co) directly into the well fluid,[49] or activation of the chlorine in the brine with an isotopic neutron source.[50]

In addition to information on fluid injection and production profiles of a well, engineers are concerned with the kind of fluid (oil, gas, or water) that is entering the well. Both neutron activation and density-measurement techniques are useful in obtaining this information. In a novel approach devised by Sutton and Albright,[51] a radioisotope-tagged oil, often crude from the same well, is pushed by suitable pumping equipment back into the formation of interest in a producing oil well. Successive runs with a gamma-ray log indicate when all the tagged oil has been displaced from the wellbore. The well is then returned to production, and the tagged oil returns to the wellbore and is removed by either flowing or pumping. Zones that are producing water retain the tagged oil for a much longer time than the oil-producing zones because of the immiscibility of oil and water. Thus successive gamma-ray logs made during the production period following the injection show the water-producing zones as radioactive areas, the ones remaining radioactive longest being those in which water saturation is highest. In many ways this work is an extension of an earlier method of Widmyer.[52]

Neutron Sources and Applications

Several developments in the nuclear field over the past 5 or 6 years have greatly augmented the number of neutron sources available to the petroleum industry for field use.[53] In particular, the commercial availability of the transuranium elements has been of great value in making efficient isotopic neutron sources. Small accelerators that can be put in a wellbore and produce a high neutron flux have also been developed.

ISOTOPIC SOURCES

The first neutron sources used by the petroleum industry were radium–beryllium [^{9}Be$(\gamma,n)^{8}$Be]. They are still used, primarily because AEC licensing is not required, but are extremely expensive; a 100-mc source emitting a total of about 1.5×10^{6} n/sec costs over $2000. In addition, they produce very intense gamma rays, which add greatly to the shielding problem.

Polonium, an alpha emitter, is used to fabricate very large neutron sources; as much as 2000 curies of polonium can be encapsulated in one unit to provide up to 5×10^{8} n/sec [^{9}Be$(\alpha,n)^{12}$C]. Although ^{210}Po has a half-life of only 138 days, its cost per neutron compares favorably with other sources. The gamma flux is relatively low, made up principally of the 4.45-Mev gamma from the (α,n) reaction. An AEC radioisotope license must be obtained before a polonium source may be procured or possessed.[54]

Plutonium–beryllium sources have many of the characteristics of an ideal neutron source for use in oil-well-logging operations. They may be obtained in sizes up to 10 curies, which emit about 1.7×10^{7} n/sec; have a long half-life, 24,300 years; emit relatively low levels of gamma rays in comparison to the neutron flux; and are relatively inexpensive. Disadvantages are the relatively large size, i.e., 16 g/curie; the likelihood of nonuniformity in distribution of the beryllium–plutonium compound with a resulting nonuniformity in flux pattern; and, probably most important, the difficulty of obtaining plutonium, since plutonium is fissionable and a primary ingredient of nuclear weap-

ons and has certain licensing and possession restrictions. No more than 20 curies may be shipped in one container, although it is known that 100-curie plutonium–beryllium sources are safe. In addition to requiring a special materials license, plutonium cannot be purchased but must be leased from the AEC. The fee is nominal, about $30 per curie per year, based on a charge of 4.75% per year on a value of $43 per gram of plutonium.[54]

If americium–beryllium sources were available at low cost, they would rapidly supplant other neutron sources in logging operations. They produce the lowest gamma background of any common source, emit approximately 2×10^6 n/(curie)(sec), and may be obtained with high specific activities (0.34 g of americium oxide per curie). In addition, since americium is not fissionable, it may be purchased under an ordinary radioisotope license instead of leased. Its cost to date, however, has limited its general use.

Actinium has been investigated by at least one service company in the United States in past years as a possible source of alpha particles for neutron production. A recent paper[55] describes an extensive investigation conducted in Russia for the same purpose. Both actinium–beryllium and actinium–boron sources are described. Boron produces a somewhat lower gamma flux than beryllium. Actinium-227 has a 22-year half-life and is a naturally occurring isotope, an advantage in the United States since no AEC license is required.

ACCELERATORS

Although for more than 10 years many organizations have been conducting research on the development of an accelerator for use as a neutron source in a wellbore, only a very few publications have appeared describing such units. One patent, although not issued until 1963, was filed in 1949 and describes several methods of construction of such a downhole instrument.[56] Only recently have publications appeared describing actual field operation of such devices.

An accelerator capable of producing an intense neutron flux with a relatively low accelerating voltage (low compared with most devices of this nature) almost always uses the deuterium–tritium nuclear reaction. When a deuteron is accelerated through a field of about 100,000 volts and permitted to strike a tritium atom, a neutron is emitted from the tritium nucleus with an energy of 14 Mev.

The elements required to construct such an accelerator are a source of charged deuteron particles; an evacuated beam tube with a tritium target; and a high-voltage source of 75,000 to 100,000 volts or more. Packaging such a combination of elements in a steel tube that is less than 5 in. in diameter and can withstand pressures of thousands of pounds at temperatures of several hundred degrees Fahrenheit* is not a simple task, particularly since the unit must be remotely operated through 15,000 to 20,000 ft of armored cable. Even so, a recent paper[57] describes a sealed tube capable of producing 10^{10} n/sec under steady-state conditions. Many units are designed for either pulsed or steady-state operation.[58]

One such unit is described in a paper[59] that also describes preliminary tests of the tool as a source of neutrons for downhole activation analysis, using only the prompt neutrons generated by 5-μsec neutron pulses. Youmans et al.[60] describe another instrument of this type which is used for neutron-lifetime logging. This is a new application of an old principle: determination of the capture cross section and atomic density of a substance — in this case of the rock surrounding the wellbore — by measuring the decay time of a neutron pulse. The neutron flux surviving during the period immediately preceding a neutron pulse is determined by measurement of the prompt-gamma flux produced by thermal-neutron capture.[61] Among the more common elements present in the usual oil-bearing formation, chlorine has the greatest effect on neutron lifetime. Salt water can be identified, and, using information obtained from other logging methods, formation fluid saturation can be measured.[62]

By providing both a source of neutrons that may be turned on and off quickly by electronic means and much higher neutron fluxes, possibly by several orders of magnitude, these miniature accelerators have opened an important new vista in well logging. A monoenergetic source of 14-Mev neutrons may ultimately provide even

*Subsurface temperature, °F = (depth in hundred feet)(geothermal gradient) + 74°F.

Geothermal gradient in the Texas Gulf Coast is 1.6 to 2.2°F per 100 ft. Therefore a 15,000-ft well could have a subsurface temperature of 404°F.

further advantage. The Soviet Union has produced a number of these generators and is using them for routine mineral exploration as well as for the more exotic forms of petroleum well logging.[63]

Gamma–Gamma Density Logging

Gamma–gamma density logging has been used for at least 15 years, although some investigators do not feel that it has received the attention it should.[64] The principles involved have been described by a number of writers during the past few years.[65,66] Basically the attenuation of gamma rays in both total flux and energy as they pass through a formation is measured and interpreted as a function of the density and the porosity of the formation of interest. Several isotopes have been used as sources, although ^{137}Cs continues to be the most popular because of the suitable energy of its single gamma-ray emission and its long half-life (30 years). This method, like most radiometric methods, is now in worldwide use.[66,67]

A very recently announced[68] method of determining the porosity of core samples, although not classified as a logging device, uses the same basic idea of gamma-ray interaction with the electrons within the formation matrix. The device, called a Gamma-Ray Attenuation Porosity Evaluator (GRAPE), has a drive mechanism that moves the core samples between a gamma source and detector. The porosity of the core may be calculated from the bulk density as determined by gamma attenuation. Reportedly, 100 ft of core may be analyzed per day in the field.

In a patent issued to Egan et al.,[34] an in situ method and apparatus are described to measure the location of zones or formations from which fluids are flowing into a wellbore. The differences in densities of various wellbore fluids — water, oil, and gas — are determined by a wellbore probe containing a source of penetrating gamma radiation, e.g., ^{137}Cs. Within the probe, at a fixed distance from the gamma source, is a gamma detector, electrically connected to a recording device at the well surface. The detector is shielded from direct radiation from the source but will detect any gamma rays traveling through the wellbore fluids. Because fluids absorb gamma rays in proportion to their densities, the number of gamma events detected is inversely proportional to the density of fluid surrounding the probe at any given depth. A change in the number of events detected indicates a change in the density of fluid in the wellbore. Calibration of the detector for the particular fluids expected in a wellbore will enable the logging operator to determine the kind of fluid present. When contrasting densities are correlated with the depth of the probe at any given instant, the exact location and type of fluid entry within a wellbore may be determined.

Caldwell and Sippel[69] patented an apparatus for a similar measurement, the primary difference being in refinement of the shielding section of the probe. The emitted gamma rays are highly attenuated in every direction except upward through the borehole fluids to the detector so that gamma-ray scattering by the surrounding formation is minimized. Recorded densities are therefore those of the borehole fluids and not of the surrounding formations.

Field application of such a method as covered in the two previous patents has been described. Bird and Bullard[70] reported the applications and field results obtained with a production logging defice referred to as a fluid-density analyzer. This tool also operates on the principle that the number of gamma rays reaching a detector is inversely proportional to the density of the fluid between the source and the detector. They report that the latest version of the tool will probably determine, to the second decimal point, the density of the fluid in which the tool is immersed, and they further claim that this accuracy may be maintained at temperatures from 0 to 350°F.

Reservoir Fluid Tracers

The need for a method of tracing the movement of fluids within a petroleum or gas reservoir has been recognized almost since the beginnings of the petroleum industry. Dyes were used for many years, although sorption prevented useful results except in cases involving gross fractures. The advent of radiotracers improved the situation markedly, partially because of the ability to detect these tracers at extremely low concentrations. Radiotracers also permitted the use of large carrier-to-tracer ratios, and, more important, one tracer, tritium, could be incorporated directly into the traced molecule, i.e., water or hydrocarbon. A

number of other radioactive tracers have been evaluated for oil-field use.[46,71] Only ^{131}I as the iodide seems satisfactory for a water tracer where serious sorption problems exist, but its 8-day half-life limits its utility under most circumstances. Because of the close parallelism in application of chemical and radioactive tracers, recent developments in the use of chemical tracers are mentioned briefly.

CHEMICAL WATER TRACERS

Chemical tracers are used, even in the face of the higher detection sensitivity of radioactive tracers, in several phases of petroleum-reservoir exploitation. Chemical tracers find useful application (1) when a reasonably short transit time between wells is expected; (2) when the possibility of contamination of potable water supplies exists; and (3) particularly when only one or two determinations are necessary and no radiation-detection equipment is available.

Boyd and Perry[72] reported excellent results from the use of ammonium thiocyanate as a chemical tracer for waterflood production problems. The tracer is not highly sorbed on the formation and is easily detected on location, thus requiring no laboratory analysis except with extreme dilution. Concentrations as low as 20 ppm can be detected with a visual test. They report three field tests on wells suspected of injected-water bypassing.

Organic chemical tracers have been used successfully in Canada to determine segregation of production intervals in multiple-completion wells.[73] The field procedure is to introduce a chemical tracer into the higher pressure zone of a multiple-completion well and to monitor the other producing zones for its presence. If tracer is detected, communication exists between the zones and remedial measures can be initiated to isolate the producing intervals. The chemical tracers field tested were phenolphthalein-alcohol solutions in crude oil or brine, phenolphthalein in dimethylsulfoxide and ethylene glycol solution, and diethyl yellow (*p*-diethylaminoazobenzene) and phenolphthalein in wax-stick markers.

Brigham and Smith[74] report results of a chemical tracer test using ammonium thiocyanate and potassium iodide to evaluate a five-spot waterflood pattern. They were more interested in analyzing the tracer behavior than in developing tracer techniques or field testing chemical tracers, and they developed equations to predict tracer breakthrough and peak concentration as well as a general equation of the breakthrough curve. This theoretical analysis of tracer behavior should prove most useful in evaluating field-test results.

RADIOACTIVE WATER TRACERS

In general, emphasis on radioactive fluid tracers has been on improvement of existing methods and elimination of the less workable ones rather than on totally new developments. Tritium has emerged as a more or less universal oil-field fluid tracer.

Heemstra, Watkins, and Armstrong[71] evaluated nine radionuclides for waterflood tracer application. Results of laboratory flow tests through sandstone cores showed that three of the materials tested should be useful as water tracers: tritiated water, ^{131}I, and ^{124}Sb.

Greenkorn[75] conducted laboratory and field tests on 13 possible waterflood tracers, all chemical except tritiated water, in an effort to find five that could be used together in evaluating a planned pilot flood. The tritiated water showed excellent tracer qualities.

The application of radioactive tracers in evaluating miscible-phase production operations has recently been described.[76] Tritium was used to trace the flow of injected gasoline in one experiment and of isopropanol and water in another. Of interest in the gasoline experiment was the use of tracers to determine the relative vertical distribution of the injected fluids within the producing zone by determining the radioactivity of the liquids extracted from cores obtained from the flooded zone during the test.

Many other applications of tracer studies in specific oil fields have been made, although the results are not published. For example, a major oil company in the United States used over 200 curies of tritium last year in routine field tracer studies, although no publications appeared. In addition to the evidence presented by increasing purchases of tritium by oil companies, the actual number of tests being made with a given amount of tracer is increasing because of successful efforts to increase detection sensitivity and thus decrease the amount of tracer required per test.[77]

The basic high sensitivity of the radiation-detection method may be achieved without use

of any radioactive material in the field system by a recently patented technique[78] in which neutron activation analysis is the detection method. A stable tracer substance, such as iodine, is added to the injected fluid to be traced, the tracer is made radioactive at the point of detection, and the radiation produced is measured quantitatively by common radiometric techniques. A neutron source producing a flux of at least 10^7 n/(cm^2)(sec) is used for the activation. The method has application in tracing either liquids or gases.

RADIOACTIVE GAS TRACERS

The use of tritium and ^{85}Kr as tracers for natural gas and other gases injected into natural-gas reservoirs has proved particularly fruitful. Krypton-85 has an advantage over tritium because of the greater ease of detection of its energetic beta radiation.[79] In addition to actual well and reservoir operations, techniques have been devised for flow measurements in complex gas-flow systems.[80,81] Krypton-85 was used as a gas tracer in the huge Spraberry field of West Texas to investigate directional orientation of fractures. Results obtained from tracer breakthrough indicated a fracture trend that agreed well with predictions made from core-orientation studies.[82]

A unique application of dual tracer injection for tertiary-recovery evaluation was recently made in the Appalachian area. Subsurface gas-tracer tests were conducted in conjunction with an in situ combustion field test. Krypton-85 and tritium were selected as the radioactive tracers with helium as an auxiliary tracer. The ^{85}Kr determined the transit time between the injection and production wells; the tritium tracer indicated vertical distribution and efficiency of the combustion zone; and the helium indicated sweep efficiency in the area.[83]

A similar test conducted in the same general area used ^{85}Kr to determine transit times between wells, the presence of highly permeable streaks within the producing zone, and the relative efficiency of the injection gas.[84,85]

The use of multiple radiotracers in secondary-recovery and underground gas-storage operations is sometimes advantageous in the evaluation of a particular process of the operation. Determination of two or more radioisotopes in a single sample presented several problems; however, especially when accurate quantitative data were required, Lewis[86] gave a method for the radiochemical determination in natural gas of tritiated methane and ^{85}Kr—two tracers commonly used in combination. The method comprises oxidation of the gas, volatilization of the components in a vacuum, and counting the activity of the individual isotopes. It is also applicable to several other isotopes and compounds, e.g., ^{14}C, ^{37}Ar, and almost all tritiated hydrocarbons.

Clewell[87] has a method for determining the efficiency of a flame front during in situ combustion. He proposes using tritium as a tracer and analyzing the produced fluids for tritium content, either as gaseous tritium or tritiated water. The presence of tritium gas in the produced fluids indicates inefficient combustion of the injected gases. The tritiated water produced indicates that the combustion front is close to the producing well.

Miscellaneous Fluid-Tracer Uses

In addition to actual reservoir fluid-flow evaluation and measurement, radioactive tracers are extremely useful in drilling and completion operations. One of the earliest industrial applications of radioactive tracers was the use of carnotite as a tracer in oil-well cementing to determine the position and homogeneity of cement behind the casing. The long half-life of the radium present in the carnotite is extremely undesirable in this application because of interference with possible subsequent radioactivity measurements in the well. Today, operators use ^{131}I or other short-half-life tracers for this purpose.[88]

Radioisotopic methods have been used in drilling operations to determine many kinds of data previously poorly known or unobtainable. For example, a method has been proposed to control the circulation of drilling fluid by post-radioactivation techniques based on the assumption that, during normal drilling operations, the ratio of drilling fluid entering and leaving the wellbore remains relatively constant. Any deviation in this ratio indicates a potentially hazardous condition. The first indication of blowout usually is when the fluid output greatly exceeds the input. Lost-circulation problems are present when the reverse is true. This method requires either a neutron- or a gamma-radiation source and detectors attached to the drilling-fluid output and input flow lines. An element

within the drilling fluid is made radioactive and detected. The amount of radioactivity detected is dependent, among other things, on the flow rate. The automatic comparison of the radioactivity detected in the two flow streams provides a means of monitoring the fluid-circulation rate.[89]

The invasion of cores, cut with water-based muds, by the mud filtrate has always presented a problem in interpretation of fluid-saturation data. A method developed by Armstrong and Lovelace[90] uses tritiated water as a tracer in the mud system to determine quantitatively the extent of such invasion. At least one oil-field service company in the United States now offers it as a routine service.

Hydraulic fracturing of oil-bearing formations has become a very important completion technique.* Szymanski has used small spheres tagged with 40 μc of ^{59}Fe to mark the location of perforations made as initiation points for fractures.[91] He also points out that the use of tagged sand in the fracturing process produces data that are difficult to evaluate. This fact was recognized by Mihran, of the Halliburton Co., who devised a very complex system of data evaluation.[92]

Conclusions

In summary, it should be emphasized that this report is by no means all-inclusive; space does not permit total coverage of all the work being done today with radioisotopes in petroleum and natural-gas reservoir exploitation. However, a representative cross section has been presented. It is evident that the period of exploratory research has ended and that a firm basis of industrial application has been established. Developments during the past 5 years have been definitive in nature and have reduced many experimental applications to accepted practice.

This practice may be expected to continue in the future, and many of the newer applications that now are experimental will either become routine or be abandoned. Obviously many applications have been developed with an Edisonian approach, and many of the data so obtained are applied empirically. The theoretical aspects of these practices should receive attention in the immediate future, and greater utility should result. This is particularly true of the use of radioactive tracers in the study of fluid dynamics of petroleum reservoirs. Although a large body of literature has been published in this area, only a few papers deal with the theoretical analysis of tracer-obtained data. Much research remains to be done. Additional references[91–121] are given for those who wish to dig deeper.

*Completion technique is basically the last operation performed on a new well to prepare it for production or injection; this might include such techniques as hydraulic fracturing, shooting the well with nitroglycerin, acidizing, or perforating.

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Section III

Isotopes and Radiation Technology

Process Radiation Development

Technology in the production of wood–plastic combinations (WPC) is developing rapidly. Two companies, American Novawood Corporation and the Lockheed-Georgia Company, are already commercially producing developmental quantities of WPC. Other companies, although not actually in production, are sponsoring some of their own development work; however, since their successes are apt to be of a proprietary nature, they probably will not appear in the open literature—at least for the present.

In this section, a literature review—the second in a series on radiation polymerization—emphasizes technical aspects in the production of WPC. This is followed by a summary of an economic and product evaluation, issued by Arthur D. Little, Inc. Although this 1964 report was prepared from the most recent data that were then available, the state of the art has advanced considerably since that time. Nevertheless, this report does present some of the fundamental technical and commercialization problems and gives, as well, both a base line from which more recent developments can be projected and some ideas as to the products for which WPC might be commercially profitable.

Finally, there are presented some highlights of an Atomic Industrial Forum–Atomic Energy Commission (AEC) meeting on the present status and future potential of WPC.

Recent Developments in the Production of Wood–Plastic Combinations

This article, the second in a series on radiation polymerization, is a literature review emphasizing technical aspects of producing wood–plastic combinations (WPC)—action of radiations, inhibition and acceleration of polymerization, and physical parameters. These materials can be produced by several methods. Conventional techniques use chemical catalysts to initiate polymerization. Radiation techniques involve monomer impregnation, with or without prior swelling of the wood, and irradiation. Early and recent experiments at West Virginia University which are reviewed include determination of optimal conditions for producing WPC, addition of chemicals to speed up polymerization, dose-rate studies, copolymerization, production of colored WPC, and further tests on physical properties of the products. Selected literature on the effects of radiation on wood components and on monomers is also reviewed.

The subject of radiation-treated impregnated wood, or wood–plastic combinations (WPC), has been discussed by authorities in several fields. Most recently *Chemical Engineering*[1] reported on the present status of WPC, summarized briefly an Arthur D. Little, Inc., report[2] on the technical and economic aspects of WPC, and touched on the possible significance of WPC to the chemical processing industries. Among the regional inquiries[3] into commercial feasibility* that have been launched are those by Battelle Memorial Institute, Western New York Nuclear Center, and the Southern Interstate Nuclear Board (SINB). The potential commercial applications of radiation-induced WPC in the Southeast, Northwest, and Northeast were discussed at a recent meeting in Chicago.[4] North Carolina State of the University of North Carolina at Raleigh is evaluating Appalachian region woods and WPC.[3] Machurek, Dietz, and Stein[5] outlined typical properties and uses of WPC in 1964 at Geneva in a paper on chemical processing by radiation. In earlier issues of *Isotopes and Radiation Technology*, Fowler and Aebersold[6] defined the place of WPC in relation to the total U. S. program on isotopes and ra-

*The name "Novawood," used for a time for WPC, has been dropped; Lockheed-Georgia Company calls its product "Lockwood."[3]

diation development, and Ballantine[7] reviewed the work up to 1964 at West Virginia University. Elsewhere in this section of the present issue, the Arthur D. Little study is discussed.

Industry has shown considerable interest* in WPC developments,[8] and news of these products has been appearing often in the press and in trade journals,[9] stimulated partly by traveling exhibits of WPC products in the South and East. In cooperation with SINB and the Southern Furniture Manufacturers' Association (SFMA), Stanley Furniture Co., Stanleytown, Va., made several articles from WPC for display in 1965 at retail stores and other establishments. This company claims to be the first to make WPC into furniture for use. One Stanley officer has been named to a SFMA committee on site selection for WPC plants, and another officer represents SFMA on the 17-state SINB. In other developments a sample of WPC flooring, formerly shown at the Smithsonian Institution's Museum of History and Technology, was installed at the Federal Pavilion of the New York World's Fair. The 16- by 16-ft parquet section, made by Lockheed-Georgia Company, was impregnated with methyl methacrylate and polymerized with ^{60}Co. A WPC household-utensils exhibit also has been prepared by American Novawood Corp., Lynchburg, Va.

Progress in WPC was reported in 1962 to workers in radiation-induced polymerization and graft copolymerization[10] and in 1964 to those interested in nuclear applications[11,12] to the wood, paper, and pulp industry. Russian workers[13] wrote, as early as 1960, of the application of WPC in the metal-casting process, predicted commercial polymerization of monomers with textiles and paper, and mentioned the possibility of preserving fibrous museum items and archaeological treasures by the WPC method. In a brief, unsuccessful attempt to improve the moisture absorption and dimensional stability of wood, Ballantine and Manowitz[14] in 1956 found that methyl methacrylate could be polymerized in birchwood dowels at a total dose of 4.5 Mrep.

Technical work on WPC has been sponsored by the Division of Isotopes Development (DID) primarily at West Virginia University. Perhaps indicative of the state of the art, DID contracts recently were awarded to Lockheed-Georgia Company, Vitro Engineering Co., and Research Triangle Institute. Lockheed-Georgia, which produces a WPC material known as Lockwood, is processing samples submitted by 78 manufacturers. Vitro Engineering is executing a conceptual design of a WPC pilot plant. Research Triangle is investigating physical and mechanical properties of irradiated specimens being prepared at West Virginia University, including southern pulpwoods, such as loblolly pine. A number of universities and laboratories have studied WPC: McMaster University (Canada) and Atomic Energy of Canada Ltd. (AECL), the State University College of Forestry at Syracuse University (N. Y.), and Dow Chemical Co.

In the present article, the second in a series on radiation polymerization,[15] recent West Virginia University work, patents,* and selected literature are discussed. Emphasis is placed on technical aspects of WPC — mode of action of irradiation, competitive methods, inhibition of the polymerization, and control of the WPC process — rather than on commercial or economic aspects.

Production of WPC is among the attractive radiation-induced reactions (Table III-1),[16] although its relative position in this list of reactions or among others being studied may be debatable. According to *Nucleonics Week*,[17] radiation-induced chemical reactions may be a more fruitful nuclear pursuit than the graft polymerization of vinyl compounds on cellulose with

Table III-1 SOME CURRENTLY ATTRACTIVE RADIATION-INDUCED REACTIONS

Reaction	G value*
$CO + C_2H_4 \rightarrow$ copolymer	$10^4 - 10^5$
$C_6H_6 + H_2O + O_2 \rightarrow$ phenol	50
Alkane + $SO_2 + O_2 \rightarrow$ sulfoxide	$\sim 10^3$
Alkane + $SO_2 + Cl_2 \rightarrow$ sulfonylchloride	$10^5 - 10^6$
Alcohol + $C_2H_4 \rightarrow C_{12}$ to C_{20} alcohols	$\sim 10^3$
Alkyl halide + tin $\rightarrow R_2SnX_2$	$\sim 10^2$
Wood + monomer → WPC	High
Wood + coating → cured protective coating	High

*The G value is the number of molecules formed or reactant consumed per 100 ev absorbed in the reaction system.

*In an invitation by AEC to bid on a conceptual design study of a large-scale WPC plant, 28 firms responded[8] by Aug. 31, 1964.

*Besides the patents mentioned in this paper, the AEC has on file two patent applications for the uniform dyeing and fire retardation of WPC.

gamma rays. This view was based on remarks[18] by J. W. Hodgins, McMaster University, who used a swelling technique to produce WPC. The weakening effect of the water needed to swell the wood and provide grafting sites apparently offset the strengthening characteristics of the graft copolymer in Hodgins's experiments.

Wood–plastic combinations with improved physical properties have been obtained by other workers without the use of water as a swelling agent. The ratio of homopolymer to graft copolymer in the wood has not been precisely determined, but extraction of plastic from WPC with organic solvents indicates that graft copolymerization of the plastic with cellulose accounts for only a small percentage of total polymerization in the wood.

According to Davies,[19] limited work at the Commercial Products Laboratories of AECL is now concerned with techniques of impregnation and irradiation of samples. This laboratory plans to examine the chemistry of WPC on a modest scale and to obtain data for a small pilot plant to treat Canadian wood.

Grafting of Polystyrene

Perhaps indicative of the rapidly changing nature of WPC development, Ramalingam, Werezak, and Hodgins,[20] at the Fourth Cellulose Conference at Syracuse University in 1962, reported substantially enhanced bending strength and dimensional stability in the grafting of polystyrene to the cellulose in red pine sapwood. Flammability of the material was noted as a serious deterrent to its use in structures, and experiments with halogenated styrenes were planned. (Air Reduction Company, Incorporated, later reported data[2] on the effect of polyvinyl chloride on the combustion properties of WPC.) Polystyrene-cellulose grafting was accomplished by gamma irradiation of a ternary solution of styrene, methanol, and water. Optimum conditions,[20] determined by a factorial experiment, are

Evacuation time (25°C, 1 μ), hr	11.5
Impregnant composition, wt.%	
Styrene	53
Methanol	43
Water	4
Impregnant uptake, %	125
Irradiation dosage, Mrads	4
Heating temperature, °C	75
Heating time, hr	9
Average enhancement of bending strength, %	55

The wood samples were irradiated in a swimming-pool reactor. Dose rate was about 1 Mrad/hr; less than 5 Mrads was given per sample. After irradiation the wood was placed in an aluminum pressure canister and the temperature elevated to about 85 to 105°C for periods of up to 24 hr to complete the polymerization.

Electron-spin resonance (ESR) studies indicated the existence of two distinct types of long-lived free radicals, one of which was believed to be the specific precursor in the grafting reaction. The maximum radical concentration, as indicated by ESR, was about 2×10^{18} per gram. If the molecular weight of cellulose is 150,000 to 300,000 (1500 to 3000 cellulose units per chain), this corresponds to one or two free radicals per cellulose molecular unit. This number suggests to these authors that the unpaired electron is located at the end of the molecule. From other ESR experiments it was concluded that the free radicals arise from the wood, not from the methanol, styrene, or water.

The effect of heating on free-radical formation also was noted (Fig. III-1). The drop in free-radical concentration was attributed to formation of graft polymer. For wood samples in vacuum, however, this initial sharp decay did not result, and the radical concentration decreased very slightly over several days.

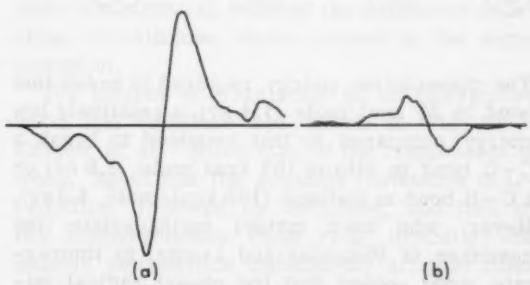
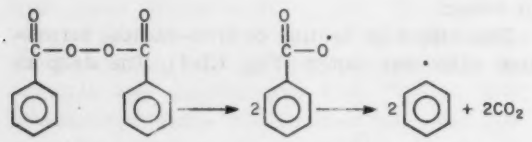


Fig. III-1 Effect of heating on free-radical concentration[20] as shown by ESR spectrum (a) immediately after irradiation and (b) after 2-hr heating at 105°C.

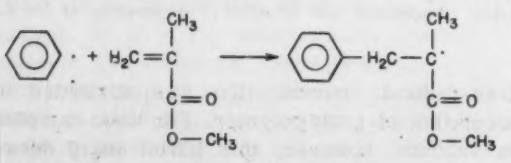
The improvement in bending strength for wood containing graft copolymer was of the same order as that observed for wood containing homopolymerized styrene. Copolymerization gave advantages of better physical appearance and insolubility in aromatic solvents.

Radiation- and Catalyst-Induced WPC

Irradiation should be balanced against other methods in the production of WPC. In one instance, Meyer,[21] after discussing radiation-induced WPC with furniture manufacturers in western New York, reported little interest in building and operating an irradiation facility at their plants, and transportation of wood products was noted as one serious deterrent to the acceptance of the radiation method. It should be noted, however, that production of WPC by any method would entail another step in wood processing. Observing that Erickson and Balatinecz[22] used peroxide and heat polymerization of styrene in wood to study liquid flow paths, Meyer stated that the addition polymerization reaction is the same regardless of the source of free radicals. (Free-radical formation by irradiation has been described earlier.[15]) Meyer used the heat decomposition of organic benzoyl peroxide as a source of free radicals:



The dissociation energy required to break this bond is 35 kcal/mole (1.4 ev), a relatively low energy compared to that required to break a C–C bond in ethane (83 kcal/mole, 3.6 ev) or a C–H bond in methane (100 kcal/mole, 4.2 ev). Meyer, who used methyl methacrylate (the monomer of Plexiglas and Lucite) to impregnate wood, noted that the phenyl radical initiates polymerization:



Impregnated samples of straight-grained yellow-birch sapwood received one of three treatments: (1) irradiation with a 1000-curie source (5 Mrads), (2) addition of 0.2% benzoyl peroxide and direct heat, or (3) addition of 0.2% benzoyl peroxide and radio-frequency heat. The apparatus, which resembles that used in radiation polymerization of WPC, is shown in Fig. III-2. Physical properties of treated and untreated samples differed, but no significant difference was seen in the hardness or compression of WPC produced by any one of the three polymerization methods. Preliminary examination showed that the polymer was located in the vessels, rays, and cell lumens of the wood.

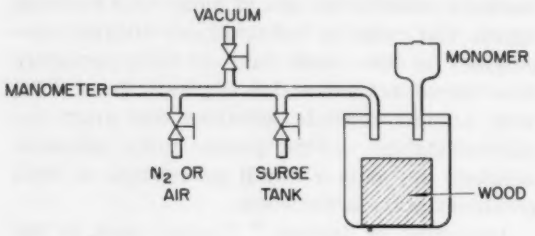


Fig. III-2 Impregnation system used by Meyer.[21]

Most reviewers of wood stabilization have mentioned the work of the U. S. Department of Agriculture's Forest Products Laboratory in cooperation with the University of Wisconsin. This laboratory has worked on chemical modification of wood since early 1940, and a bibliography of their publications is available.[23]

Five Important Aspects of the Radiation Technique

Siau, Meyer, and Skaar,[24] recently reviewed five basic aspects of the irradiation technique: (1) initiation of polymerization by irradiation, (2) effect of radiation on cellulose, (3) effect of radiation on wood, (4) grafting polymers to cellulose, and (5) monomer impregnation and irradiation of wood. Even though not all these factors are of equal concern to large-scale commercial processes, information obtained from their study is of fundamental interest and also may pertain to process efficiency.

Radiation-induced polymerization may have been observed before the discovery of X rays (1898). By 1938 Hopwood and Phillips[25] had found that styrene and vinyl acetate could be polymerized at room temperature by alpha, beta, or gamma rays "in conjunction with neutrons or alone." Various workers report that the rate of polymerization, R_p, varies with the 0.5 power, the 0.85 power, or as a linear function of the radiation intensity (Table III-2). At high intensities, one authority remarks, the

Table III-2 EFFECT OF RADIATION ON INITIATION OF POLYMERIZATION AND CROSS-LINKING PROCESSES*

Dose rate, Mrads/hr	Material	Result†	Reference, year
1.3	Bulk styrene	$R_p = kI$; 1.4% of polymerization per Mrad between 0 and 8 Mrads	24*a* (1963)
0.2	Bulk styrene	$R_p = I^{0.85}$	24*b* (1960)
0.0006 to 0.002	Styrene, methyl methacrylate in bulk, additives	Rates varied depending on additives	24*c* (1962)
	Vinyl acetate, methyl methacrylate, cellulose derivatives in aqueous solution	$R_p = I^{0.5}$; little cross-linking at low viscosities	24*d* (1953)
	Polystyrene	Cross-linking of polystyrene with high-energy radiation	24*e* (1960)
High	Polyethylene	Trapped radicals formed; scission to cross-linking ratio, 0.35	24*f* (1963)
	Cellulosics	Cross-linking with high-energy electrons, none with ^{60}Co	24*g* (1960–1961)

*Compilation was prepared from the review of Siau et al.,[24] 1965.
†The polymerization rate = R_p.

rate of polymerization decreases because of chain termination and cross-linking of free radicals.[26]

Cellulose and wood likewise were studied in early experiments, but irradiation of these materials by relatively modern techniques began around 1950. Lawton et al.[27] in 1951 irradiated wood with high-velocity electrons from a modified X-ray unit and accumulated an exposure of 0.14×10^6 equivalent r/sec (about 500 Mr/hr). Results on the irradiation of pure cellulose were reported the next year.[24h] Lawton[27] remarks:

> Irradiation of basswood with high-velocity electrons alters its structure in such a way that some of the insoluble carbohydrate components become available to rumen bacteria. . . . The cellulose in the untreated wood appears to be relatively unaffected by the enzymatic action of the rumen microbes, and the digestibility is not much increased by irradiation up to 6.5×10^6 r. Between 6.5×10^6 and 1.0×10^8 r increased irradiation results in increased fermentability.

He noted that at doses above 100 Mr the wood becomes hygroscopic and friable and surmises that carbohydrates in the wood are converted to other indigestible (to bacteria) compounds. Above 330 Mr, cellulose was completely converted to soluble material. The work reviewed by Siau, Meyer, and Skaar,[24] on wood irradiation (Table III-3) showed that many of Lawton's observations were remarkably accurate and some of them—for example, on hygroscopicity at high doses—were followed up by later workers. Mater[28] also studied the effect of high-energy radiation on wood, Loos[29] studied the effect of gamma radiation on wood toughness, and Breger, Carroll, and Bolt[30] studied the effects of radiation on wood along with coal and explosives. Smith and Mixer[31] found that ^{60}Co radiation did not affect the gross analytical composition of redwood and that the naturally occurring aromatic compounds (lignin and other extractives) reduced the number of scissions of cellulose chains caused by the same radiation.

The effects of irradiation on unimpregnated wood and cellulose are shown in Fig. III-3. Karpov et al.[13] noted that, of the components of wood, lignin has the greatest resistance to irradiation, followed by cellulose, and then by the hemicelluloses. Dose rate, intensity, and sample composition undoubtedly all play roles in the variation among these results. Certainly these factors are of considerable importance to process efficiency and economy in the production of WPC.

Extensive work has been done on the grafting of monomers to cellulose by radiation (Table III-4). Ultraviolet, as well as beta and gamma radiation, has been used. Some of this work has been mentioned previously.[15] Siau et al.[24] note that unanswered questions still remain concerning the comparative effect of electron and gamma radiation and on the combined effect of thermal treatment and ionizing radiation. For example, gamma radiation appears to be more efficient than high-energy electron radiation at high dose rates for initiation of polymerization; but for unknown reasons beta

Table III-3 EFFECT OF RADIATION ON CELLULOSE AND WOOD*

Dose	Material	Result	Reference, year
<0.1 to 500 Mrads at 0.14 Mrad/sec	Pure cellulose	Little degradation below 0.1 Mrad, degradation above 1 Mrad, complete conversion to soluble material at 500 Mrads	24*h* (1952)
0 to 500 Mrads	Wood	Decrease in mechanical strength at 1 Mrad/hr; see Fig. III-3	13 (1960)
1 to 330 Mrads	Wood	Moderate degradation below 1 Mrad (high-energy beta); entirely soluble at 330 Mrads	26 (1951)
100 Mrads	Wood	Decrease in hygroscopicity	10, 24 (1962)
1 to 100 Mrads (^{60}Co, 2- and 7-Mev electrons, fuel elements)	Cellulose cotton	No visible changes below 10^6 rads; fibrillate nature lost at higher doses; gases produced at 100 Mrads analyzed; degree of polymerization ~ total dose	24*i* (1962)
0.1 to 100 Mr	Cellulose	Degradation depends on total radiation dose; independent of dose rate; see Fig. III-3	24*a* (1963)
100 Mrads	Wood	See Fig. III-3	20, 32, 33 (1960, 1963)
40 Mrads	Wood	Minimum hygroscopicity at high doses	24*i* (1962)
Up to 10 Mrads (2-Mev electrons)	Cellulose	Relative degree of polymerization decreases with dose: 0, 1100; 1 Mrad, 700; 5 Mrads, 200; 10 Mrads, 100	24*j* (1963)
0.0001 to 10 Mrads	Wood	Decay resistance below 3 Mrads; decrease in hygroscopicity between 0.0001 and 10 Mrads	10, 24 (1962)
0.0001 to 1 Mrad	Wood	High probability of decreased hygroscopicity, swelling, and susceptibility to decay	24*k* (1959)
>1 Mrad	Wood	Degradation and increase in hygroscopicity	Several workers; see 24 (1965)
	Wood	Degradation independent of dose rate	24*l* (1959)

*Compilation was prepared from the review of Siau et al.,[24] 1965.

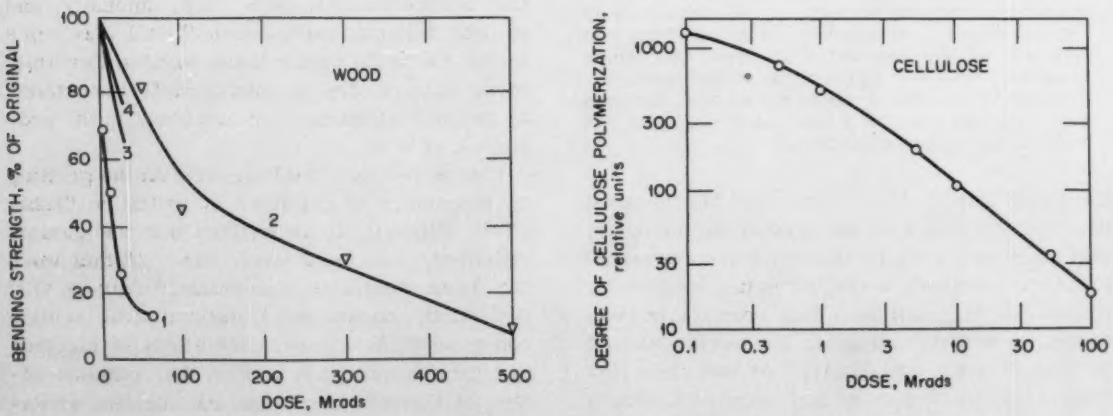


Fig. III-3 Degradation of wood and cellulose by gamma irradiation. Data are from the review by Siau et al.[24]: the wood data are plotted from data of (1) Krasnovitskaya,[32] who used a dose rate of 1.8 Mrads/hr, (2) Karpov et al.,[13] who used a dose rate of 1 Mrad/hr, (3) Ramalingam et al.,[20] and (4) Siau, Meyer, and Davidson;[33] the cellulose data are based on data by Huang et al.[24a]

Table III-4 RADIATION GRAFTING OF MONOMERS TO WOOD*

Dose	Type of wood	Monomer	Result	Reference, year
0.0001 to 10 Mrads from 1 sec to 24 hr		Several	Patents	24*m*, 46 (1958, 1963)
1 Mrad/hr (1-Mw reactor)	Red pine	Styrene	Best results at 4 Mrads	20 (1963)
0.8 Mrad/hr	Pine	Styrene	Increased in bending strength and compression at ~15 Mrads	13 (1960)
0.001 to 0.4 Mrad/hr (^{60}Co and 2-Mev Van de Graaff)	Ponderosa pine sapwood	Styrene	Graft polymerization; polymer loading up to 90%; antishrink efficiencies up to 70%; mechanical strength not appreciably changed	24*n* (1962)
0.055 Mrad/hr	Many species	Methyl methacrylate, vinyl acetate, ethyl acrylate, styrene, ethylene glycol, dimethylacrylate	Considerable increase in weight, hardness, and strength	24*o* (1962)
0.0005 Mrad/sec to doses of 40 Mr	Birch veneer	Styrene, acrylonitrile	Styrene graft copolymer determined	32 (1960)

*Compiled from the review of Siau et al.,[24] 1965.

radiation produces the highest antishrink efficiency for a given loading of polymer. J. A. Kent* suggested that these effects might be explained by heat-treatment since (1) the antishrink efficiency of wood can be improved by heating and (2) beta radiation is usually applied at high dose rates and involves heating. No comparative tests have been reported of beta and gamma radiation under similar conditions of dose, environment, and wood to determine whether or not the type of radiation produces different effects.† A similar situation exists for the combination of thermal treatment and ionizing radiation, but Siau et al.[24] believe that the two factors may interact to improve the dimensional stability of wood. No quantitative information is available on a possible combined effect.

West Virginia University

Ballantine's 1963 review[7] of West Virginia University's work on the use of gamma radiation to produce WPC was based entirely on two AEC reports,[24c,34] one of which contains an extensive bibliography.[34] Earlier and later work at this location has been presented at various conferences.[11,35–40]

EARLY EXPERIMENTS ON RADIATION EFFECTS ON WOOD AND RADIATION-INDUCED WPC

Harmison[41] in 1960 attempted to get more scientific coverage on gamma-irradiated WPC from Russia after an article[13] appeared in *Nucleonics*. Harmison noted that this article was the first published information concerning the effect of irradiation of wood both in the natural and in the treated state. He wrote to each institution that participated in the work and received no information.

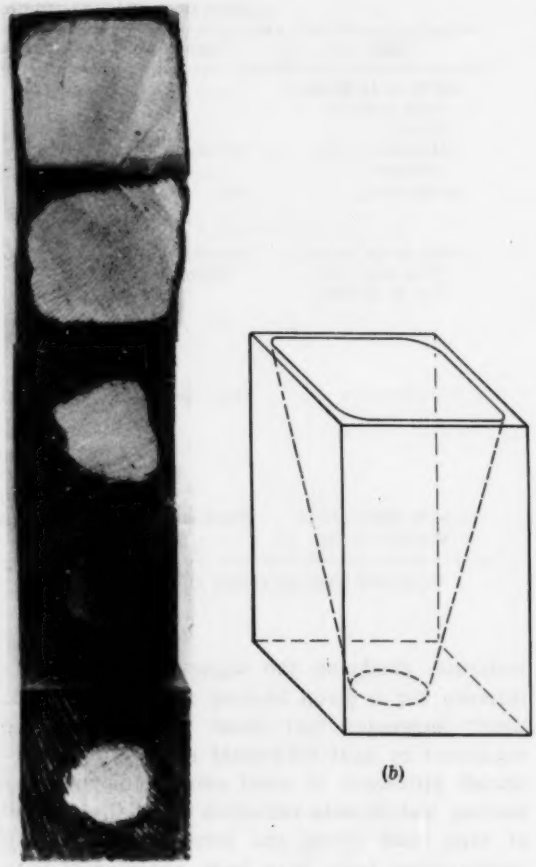
Harmison referred to two earlier, brief reports: Wakefield,[42] of Union Carbide Corpora-

*Personal communication, Oct. 6, 1965.

†According to George Adler (personal communication, Jan. 10, 1966), little work has been done on radiation polymerization with beta sources because of (1) the general difficulties in using these sources, including penetration and dosimetry, and (2) the general impression that, except for problems due to low penetration, results are not really different from those using other radiation. Whatever small differences have been found have been attributed to errors in dosimetry or to some effect resulting from the fact that the beta track length, in contrast to a gamma track length, in most experiments is much less than the thickness of the monomer layer used. Thus monomer at the surface and closer to the source will receive a relatively higher dose rate than monomer that has penetrated the wood and is farther away from the source. In addition, ionization and excitation at the end of a track are higher than at the beginning. Total dose absorbed by the entire sample thus probably is not indicative of the true kinetic picture.

tion, suggested the possible use of nuclear irradiation to avoid the embrittling effects of acid and alkali catalysts in the dimensional stabilization of wood; and McBrian,[43] of the Denver and Rio Grande Western Railroad, in preliminary studies actually irradiated creosoted crossties and found that addition of chemicals to soft woods followed by gamma irradiation induced cross-linking or polymerization. His aim was the replacement of expensive fir and oak crossties with a cheaper wood such as pine.

In his work, Harmison in 1961 studied irradiated and unirradiated samples of sugar-maple wood and irradiated samples of monomer-impregnated wood. He found that irradiated wood had a higher average elastic limit (7780 psi) compared to unirradiated wood (6460 psi) but that most of the other properties of the unirradiated wood were more desirable. On the other hand, not only was the average elastic limit increased in irradiated, impregnated wood (12,920 psi), but other properties were improved—hardness, decrease in water absorption, and dimensional stability (tangential, 3.71 times; radial, 1.68 times that of unmodified wood). He found that hardness varied with degree of polymer penetration in the wood (Fig. III-4).



(a) (b)

Fig. III-4 Variation of polymer depth in sugar-maple wood. (a) Cross-sectional view of WPC. The dark area indicates the polymer, and the center, light area the unimpregnated wood. (b) Sketch representing frustrum of unimpregnated wood in the center of block of wood. The degree of impregnation and polymerization of monomer in the wood in these early experiments was a function of Harmison's technique,[41] and, according to M. H. Stein (personal communication, Dec. 8, 1965), controlled polymer loading has been studied in greater detail.

DETERMINATION OF OPTIMAL CONDITIONS FOR PRODUCING WPC

Later work at West Virginia University dealt with determining optimal conditions and improving monomer soaking, lowering dose requirements, adding additional woods and monomers, and studying the effects of atmosphere, temperature, and dose rate. A cross section of one WPC recently produced by this group is shown in Fig. III-5.

The rate of polymerization (R_p) of methyl methacrylate ($R_p = I^{0.5}$) agreed closely with results of Chapiro[44] and was in accordance with the square-root law. Addition of carbon tetrachloride raised the slope to 0.69, but curves for unpurified monomer were considerably below those of pure monomer. The R_p of unpurified monomer was a function of the square, rather than the square root, of the dose. Furthermore, R_p was greater when polymerization was carried out in an inert atmosphere. It should be noted that conversion rates were measured during the early (0 to 20%) part of polymerization.

Experiments on the polymerization of vinyl acetate in bulk and in various woods were initially directed toward reducing the dose required for complete polymerization in wood. The effects of oxygen, type of wood, dose rate, and purity of the monomer on the rate of polymerization were studied. An example of two of these effects—air and purification of

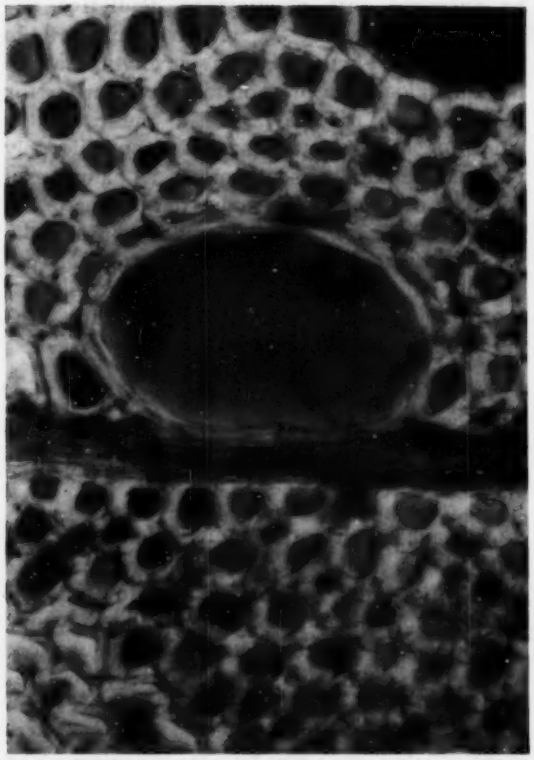


Fig. III-5 Photomicrograph of a section of dyed WPC. The wood is sugar maple, the plastic is polymethyl methacrylate, and the dye is amaplast blue (an anthraquinone dye). The smaller cells, called fiber cells, range in size from 15 to 30μ in diameter. The large cell in the center of the figure is a pore cell. The strip of cells running across the center of the figure is a ray, which does not contain polymer. The dose rate was 0.2 Mrad/hr, and the total dose was 1.6 Mrads. The gray areas indicate polymerized material. (W. E. Loos, West Virginia University, personal communication, Oct. 21, 1965).

the monomer—is shown in Fig. III-6. The bulk polymerization-rate curves showed a decrease in reaction rate and an increase in induction period with decreasing dose rate as well as an inverse relation between molecular weight and the rate of polymerization.

The log-log plot of molecular weight against dose rate (Fig. III-7) is linear with a slope of −0.45, in good agreement with the theoretical inverse-square-root relation. Although the curve lies somewhat above that of Chapiro[44] and has a slightly greater slope, the agreement is reasonably good.

Purification of monomer and the use of a nitrogen atmosphere had little effect on the

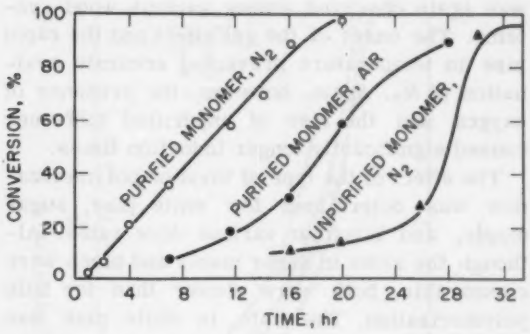


Fig. III-6 Polymerization of vinyl acetate in white pine with irradiation dose rate of 30 rads/sec. Replotted from data of Kent, Winston, and Boyle.[34] The curve for unpurified monomer in air was not available.

molecular weight of the polymer. Carbon tetrachloride, a chain transfer agent, strongly depressed the molecular weight, but this depression could not be explained on the basis of transfer alone. The striking increase in the molecular weight observed for the wood systems was explained in part by the formation of gel at higher polymerization efficiencies and in part by the slower rate of polymerization in the wood. Chain transfer with wood resins would cause the opposite effect. Depression of molecular weight caused by carbon tetrachloride

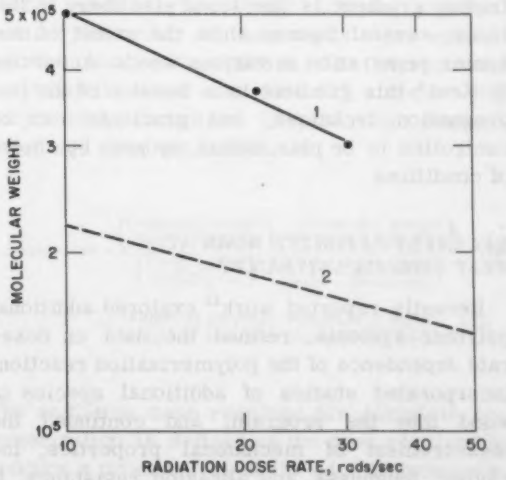


Fig. III-7 Influence of dose rate on the molecular weight of polymethyl methacrylate produced from purified monomer under nitrogen. Line 1 is a plot of the experimental results of Kent et al.,[34] and line 2 is a plot by Chapiro.[44]

was again observed among various wood systems. The onset of the gel effect and the rapid rise in temperature prevented accurate evaluation of R_p. Again, however, the presence of oxygen and the use of unpurified monomer caused significantly longer induction times.

The effect of the type of wood on polymerization was determined for white pine, sugar maple, and birch at various dose rates. Although the rates in sugar maple and birch were comparable, both were slower than for bulk polymerization. The rate in white pine was considerably slower, an effect attributed by Kent and coworkers partly to the quantity and nature of the pine-wood resins. Results with birch and sugar maple were presented in a review by Ballantine,[7] to which the reader is referred.

The total dose required to complete polymerization decreased slightly with decreasing dose rates, a finding that suggested a slightly greater efficiency of the source at lower dose rates.

Kent, Winston, and Boyle[34] found that the impregnation of hardwoods is different in many respects from the impregnation of softwoods. Capillaries, if not obstructed, provide a continuous path for the flow of the monomer. Data for the birch–methyl methacrylate system indicate that 40% of the wood is easily impregnated but that the remainder is impregnated more slowly. The so-called monomer concentration gradient is discussed elsewhere in this issue; several figures show the extent of monomer penetration in various woods. According to Kent* this gradient is a function of the impregnation technique, and gradients can be controlled to be plus, minus, or zero by choice of conditions.

RECENTLY REPORTED WORK AT WEST VIRGINIA UNIVERSITY

Recently reported work[45] explored additional polymer systems, refined the data on dose-rate dependence of the polymerization reaction, incorporated studies of additional species of wood into the program, and continued the measurement of mechanical properties, including toughness and abrasion resistance. It was found that continuous, tough radiation polymers and copolymers of vinyl chloride could be prepared with plasticizers. Radiation copolymers of styrene–acrylonitrile appeared to be feasible. Further studies involving carbon tetrachloride as a promoter or accelerator for the radiation polymerization of methyl methacrylate showed that its addition considerably reduced radiation requirements. A number of polymer dyes could be added to accentuate the wood grain and impart attractive colors to the wood. Although the dyes have inhibitory effects on the polymerization reaction, at dye concentrations that are potentially useful for WPC the inhibition is not serious.

Radiation Copolymerization of Styrene and Acrylonitrile. Although both styrene and acrylonitrile cost less than methyl methacrylate, neither monomer alone is well suited to making WPC.

Compared with polymethyl methacrylate and polyvinyl acetate, styrene–acrylonitrile copolymers offer several advantages: lower raw-material cost than methyl methacrylate, tougher and stronger polymer, less water absorption, and higher softening point than polyvinyl acetate. A feed of approximately the azeotropic composition is used in the styrene–acrylonitrile system; otherwise, a widely varying mixture of copolymers of different compositions is formed.

Part *a* of Fig. III-8 shows that there is a steady increase in the degree of polymerization per unit dose with decreasing styrene in the feed. Although the 50/50 feed ratio proceeds at the fastest rate of those reported, probably the 60/40 ratio is a better compromise between a rapid rate and a clear homogeneous copolymer.

To accelerate the rate of copolymerization of styrene and acrylonitrile, carbon tetrachloride was incorporated at various concentrations into the monomer feed. The corresponding conversion vs. total dose curves are compared in part *b* of Fig. III-8. The best activation among the various concentrations of carbon tetrachloride was 10%.

The rates of radiation polymerization of systems containing unpurified rubber-grade styrene and unpurified polymer-grade styrene were compared and found to be identical, which indicates that the cheaper rubber-grade styrene would be suitable in the process. The data show further that the purified monomer polymerizes at a rate only slightly greater than does that used "as received."

*Personal communication, Oct. 6, 1965.

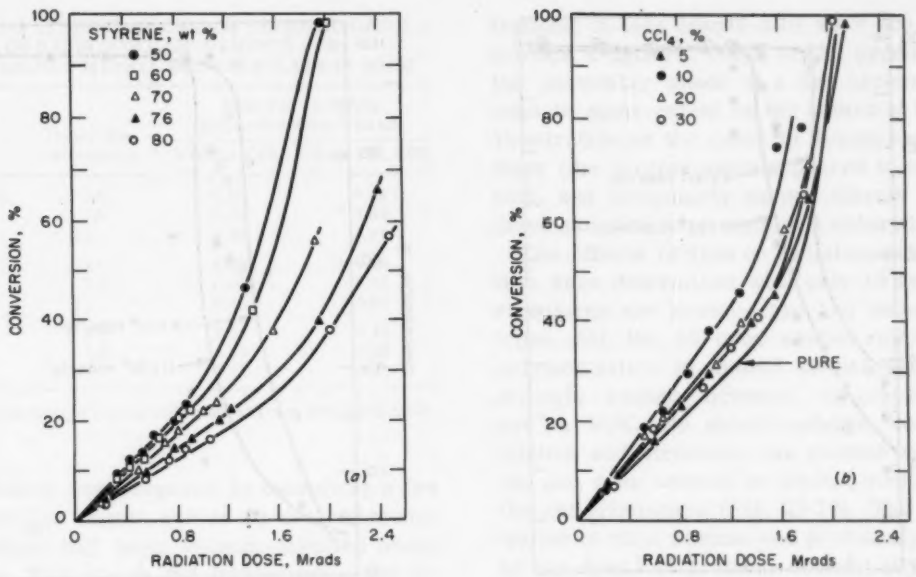


Fig. III-8 Effect on copolymerization[45] of styrene and acrylonitrile (purified monomers) of amount of (a) styrene and (b) carbon tetrachloride added to 60% styrene–40% acrylonitrile. Dose rate, 1 Mrad/hr.

Radiation Polymerization of Vinyl Chloride. Vinyl chloride, one of the cheapest commercially available monomers, can be readily polymerized by exposure to radiation. Its low boiling point (–14°C) results in handling problems; however, these appeared to Kent to be surmountable through proper engineering design. The polymer of vinyl chloride is insoluble in the monomer and precipitates out during polymerization to yield a fine, powdery, nonhomogeneous polymer with no mechanical strength. This problem was easily overcome by the use of plasticizers and copolymerizations.

Copolymerization studies suggested that 10 vol.% acrylates impart plasticity to vinyl chloride copolymers. Polymerization of vinyl chloride with 20 vol.% dioctyl phthalate and Santicizer 141* gave, in general, softer polymers than the acrylate copolymers.

The investigations with vinyl chloride did not include quantitative consideration of such factors as radiation dose requirements, conversion rates, and dose-rate dependence. Wood–plastic combinations with vinyl chloride contained fairly large but varying amounts of polymer, and the distribution of polymer throughout a given sample was very nonuniform.

DOSE-RATE DEPENDENCE OF RADIATION POLYMERIZATIONS IN WOOD

The monomers used in these tests were methyl methacrylate, vinyl acetate, styrene, and acrylonitrile. All monomers were used in the "as received" condition. The styrene and acrylonitrile were used as a 60% styrene–40% acrylonitrile solution. The "percent conversion" was obtained as follows:

$$\% \text{ conversion} = \frac{\left(\frac{\text{Oven-dry weight}}{\text{after irradiation}}\right) - \left(\frac{\text{Oven-dry weight}}{\text{before treatment}}\right)}{\left(\frac{\text{Weight after}}{\text{irradiation}}\right) - \left(\frac{\text{Oven-dry weight}}{\text{before treatment}}\right)} \times 100$$

$$= \frac{\text{Weight of polymer deposited in wood}}{\text{Weight of monomer deposited in wood}} \times 100$$

The radiation dose required for complete polymerization is defined as the dose required to produce a product free of the odor of monomer.

Radiation polymerization of methyl methacrylate in sugar maple, yellow poplar, and birch proceeded similarly (Fig. III-9). The radiation doses required to effect complete polymerization, as determined by the beginnings

*Monsanto Chemical Co.

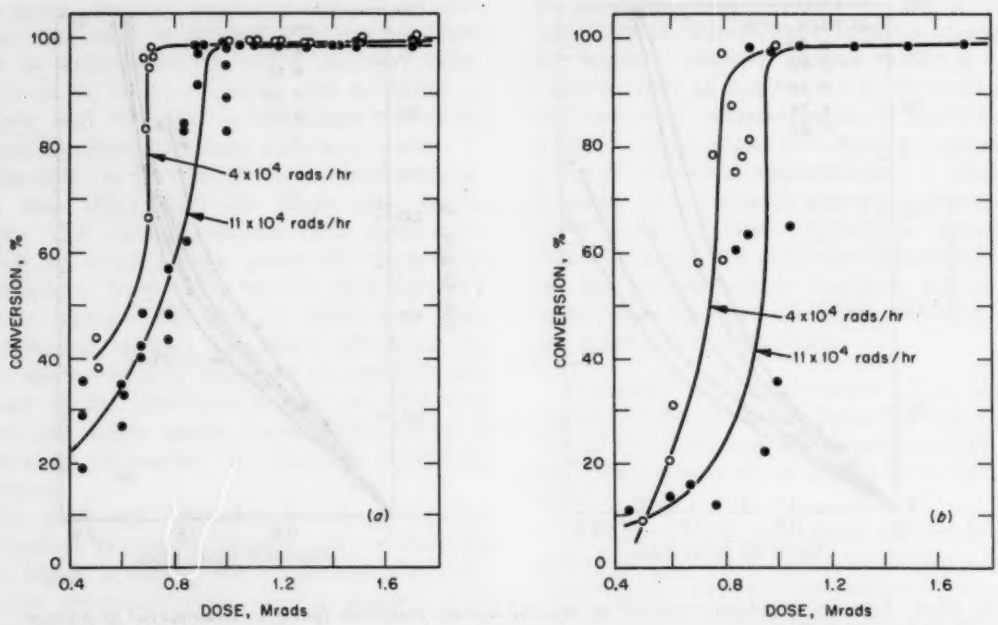


Fig. III-9 Radiation polymerization of methyl methacrylate in (a) poplar, birch, and maple and (b) white pine.[39]

of the plateaus of the respective curves, are about the same in every case and agreed well with the doses determined by an odor test.

With styrene–acrylonitrile, polymerization proceeded similarly in these three species of wood and in white pine. The total dosages for complete polymerization, as indicated by the plateaus of the curves, again agreed well with those obtained by the odor test. Within the limits of experimental error, the dose requirements with vinyl acetate are the same in the three hardwoods but different in white pine.

Polymerization of Acrylate Monomers in Maple. Polymerization of acrylate monomers impregnated in wood resulted in extensive radial splitting. Apparently oxygen plays a role in the splitting process, because the maple specimens were split when the polymerization occurred in air but not when the air was replaced by nitrogen. Later work showed that methyl acrylate but not ethyl or butyl acrylate caused splitting; Kent* believes that perhaps very rapid polymerization and excessive heating may be factors in the splitting with the use of acrylates.

*Personal communication, Oct. 6, 1965.

The wood–plastic combinations polymerized under nitrogen were hard and free of monomer and similar in appearance to those obtained with methyl methacrylate. However, since the glass-transition temperatures of the polyacrylates are far below room temperature, certain properties of wood–polyacrylate combinations would be expected to be different from those of wood–polymethyl methacrylate materials.

Effect of Carbon Tetrachloride. The effect of carbon tetrachloride on the total dose required for complete polymerization is less than predicted on the basis of bulk polymerization studies, but the effect is still significant (Table III-5).

Colored WPC. The concept that WPC products have a texture or finish that goes completely through the wood led to the idea of combining dyes with the monomers before they are placed in the wood. The dyes should be soluble enough in the monomer to provide the desired color or hues, should not appreciably inhibit or retard the polymerization, and should be reasonably stable to gamma radiation.

Table III-5 EFFECT OF CARBON TETRACHLORIDE ON RADIATION REQUIREMENTS FOR POLYMERIZING METHYL METHACRYLATE IN WOOD

Wood	Dose rate, 10^4 rads/hr	Dose for complete polymerization,* Mrads	
		Without CCl_4	With 10% CCl_4
White pine	3.6	0.50	0.45
	7.5	0.70	0.60
	11	0.85	0.70
Birch	3.6	0.50	0.35
	7.5	0.55	0.45
	11	0.80	0.60
Maple	3.6	0.50	0.45
	7.5	0.60	0.60
	11	0.75	0.70

*Based on weight loss after removal from radiation field.

A solution was prepared by dissolving a few grams of the chosen dye in the desired monomer, which had been vacuum-distilled under nitrogen. Next, using the impregnation and irradiation procedures already described, samples of the monomer–dye solution were evaluated. Finally, samples were prepared of white pine, birch, and maple and monomer–dye solution. Twenty-six dyes were found to be generally suitable with methyl methacrylate for making colored WPC. In general, dye concentrations of less than 0.02*M* were too dilute to provide a uniform color to the product. Also, the particular shade of a dye appears to depend to some extent on the nature of the wood. Penetration of the color in samples up to 3 in. thick (the largest used) appeared to be consistent, and irregularly shaped objects cut from these samples were uniformly colored.

The effects of dyes on radiation polymerization were determined with only 10 dyes whose structures are known—azo and anthraquinone types. All the 10 dyes studied inhibited the polymerization of methyl methacrylate—none strongly enough, however, to preclude their use in WPC. No direct relation between inhibition and structure was evident except that the azo dyes seemed to inhibit more than did the anthraquinones (Fig. III-10). The polymerization of vinyl acetate was profoundly inhibited by the dyes. At present, colored combinations with polyvinyl acetate will not be practical for the radiation process.

FURTHER TESTS ON THE MECHANICAL PROPERTIES OF WPC

The hardness of sugar maple–poly(styrene/acrylonitrile) increased with polymer loading

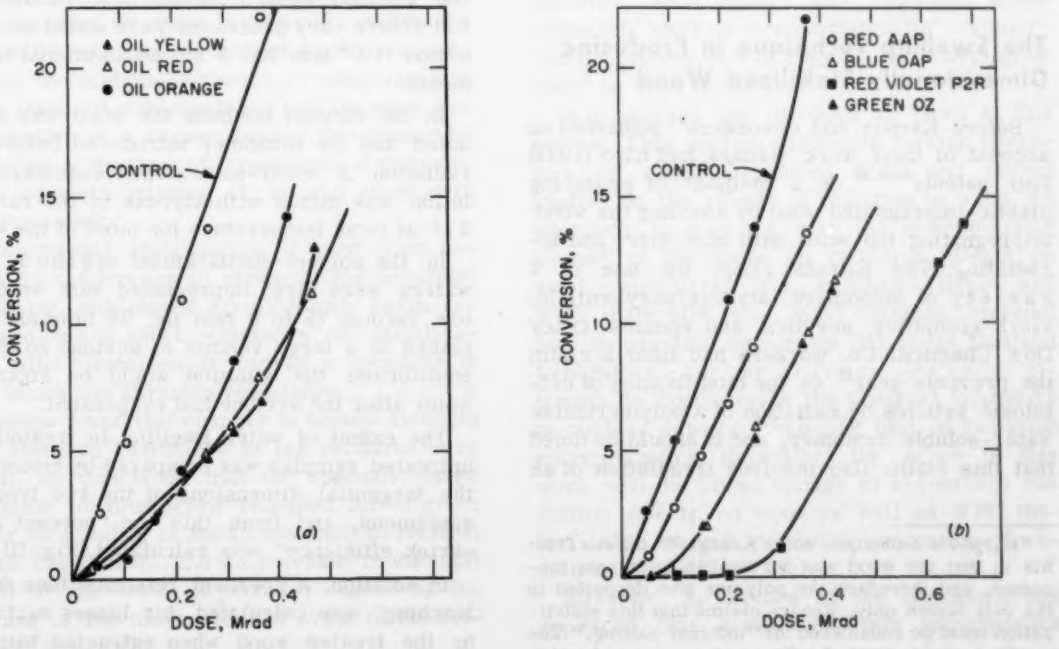


Fig. III-10 Effects[45] of (a) some azo dyes and (b) anthraquinone dyes on the radiation polymerization of methyl methacrylate. Dose rate, 10.5 × 10^4 rads/hr. Dye concentrations, 0.02M.

to a maximum value two to three times as great as the control values. At higher polymer loadings the side hardness increased with loading at a greater rate than end hardness and in some cases exceeded the end hardness.

Compression parallel to the grain for sugar maple combined with poly(styrene/acrylonitrile) was increased by as much as 50% over control values. In this series of tests it was also found that the crushing strength consistently increased over the proportional limit.

Polymethyl methacrylate improved the toughness of wood in a more consistent manner than did polyvinyl acetate. Polymethyl methacrylate increased the toughness of white pine linearly with polymer loading to about double the original value.

The abrasion resistance of sugar maple–polyvinyl acetate and –polymethyl methacrylate combinations also increased linearly with polymer loading to about three times the original (unmodified wood).

Values for static bending and shear parallel to the grain for the above-mentioned WPC were somewhat inconsistent.

Tests on glueability, weatherability, paintability, and insect resistance are still in progress.

The Swelling Technique in Producing Dimensionally Stabilized Wood

Before Karpov and coworkers[13] published an account of their work, Kenaga had filed (1958) four patents[24m,46] on a method* of producing plastic-impregnated wood by swelling the wood, impregnating the wood with monomer, and irradiating. The patents claim the use of a variety of monomers: styrene/acrylonitrile, vinyl aromatics, acrylics, and epoxies. Other Dow Chemical Co. workers had filed a claim the previous year[47] on the modification of cellulosic articles by radiation of a polymerizable water-soluble monomer, and it should be noted that this claim also involved irradiation of an aqueous solution of the monomer in the cellulosic material.

Development of the swelling-agent method for commercial exploitation has lagged behind that of the impregnation–irradiation scheme. Several variations of the techniques have been tried.

Kenaga apparently found in 1956 that, with doses below 10 Mrads, little grafting or polymerization of styrene in wood took place in relatively dry systems of styrene in solvent solutions capable of swelling the substrate. While this work was in progress, Rapson[48] and Huang[49] and coworkers reported grafting of styrene to cellulosic materials by solvent-displacement techniques followed by gamma or electron irradiation. Other workers mentioned in the Kenaga review[24n] include Paton and Hearmon,[50] Schwab, Stannett, and Hermans,[51] Chapiro and Stannett,[52] and Sakurada.[53] As early as 1956, workers at Brookhaven National Laboratory reported studies in progress on the use of water-soluble monomers, which were capable of penetrating the cellulose cell structure, and irradiation to improve the aging characteristics of wood.[53a]

Low and high vacuums, as well as solvent-displacement variations, have been used with the swelling-agent concept. Dry, immersion, and freeze–dry processes were added to evacuation (10^{-4} mm Hg, 3 hr) as additional treatments.

In the vacuum methods the wood was evacuated and the monomer introduced before irradiation. A water-saturated 1,4-dioxane solution was mixed with styrene in the ratio of 3:1 at room temperature for most of the work.

In the solvent-displacement method, wood wafers were first impregnated with water by low vacuum (5 to 6 mm Hg, 30 min) and then placed in a large volume of acetone so that at equilibrium the solution would be about 1% water after the acetone had evaporated.

The extent of water swelling in treated and untreated samples was compared by measuring the tangential dimensions of the two types of specimens, and from this the "percent antishrink efficiency" was calculated (Fig. III-11).

In addition, a "percent retention loss due to leaching" was calculated, for losses sustained by the treated wood when extracted with dioxane, as a measure of amount of homopolymer formed from controls.

*Karpov's technique, notes Kenaga,[24n] differs from his in that the wood was not swollen by styrene monomer, and therefore the polymer was deposited in the cell lumen only. Kenaga claims that this stabilization must be considered an "internal coating." The swelling-agent method allows deposition of polystyrene in the less-ordered areas of the expanded cellulosic network.

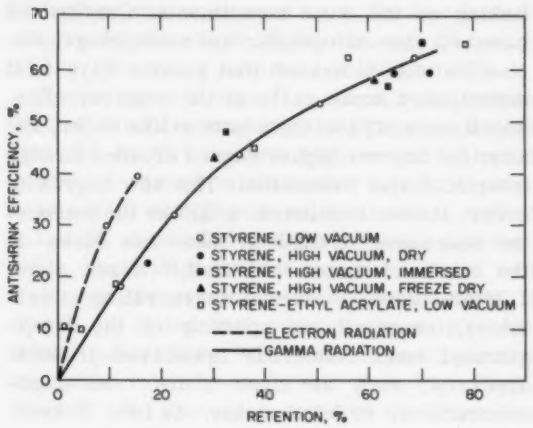


Fig. III-11 Effect of various impregnation methods, types of radiation, and kinds of monomers on the stabilization of specimens impregnated with water-saturated dioxane solutions containing monomer.[24a]

Kenaga claimed that the amount of material required for a given order of stability was relatively high by his method compared to such methods as acetylation. He attributed this partly to deposition of homopolymer in the cell lumen and mostly to grafting on the cell wall.

The solvent-displacement technique was an efficient method of impregnation. The styrene was expected to lodge completely within the cell walls by diffusion inward during evaporation of the acetone. Although no measurements were taken, it was concluded that the wood was not swollen to a degree beyond that caused by the water at the time of irradiation. A styrene–ethyl acrylate mixture (1:1) was tried with similar results.

Dimensional stabilization of wood often results in a decrease in strength; for example, treatment with phenol formaldehyde resin results in loss of shock resistance, and stabilization with formaldehyde causes a decrease in both shock and abrasion resistance.

Kenaga found that changes in tensile strength and abrasion resistance in his products were small. He concluded that the unusually large retention of monomers required for a given order of stability is due to homopolymerization in the cell lumen, and an increase in the total swell of the wood is probably caused by the opening of the more ordered areas of the cellulosic structure.

Impregnation and irradiation under high vacuum, as compared to low vacuum, had little effect on stabilization at high radiation dosages. An inhibition by oxygen was evident at low dosage. The best impregnating solution was an aqueous solution of dioxane and styrene. With ethanol–styrene solutions much homopolymerization was seen, and with an aqueous acetone–styrene solution, retentions of polystyrene were low. The solvent-displacement method of impregnation using acetone as the intermediate replacing agent gave more stabilization for a given amount of polystyrene. Electron irradiation of specimens impregnated with an aqueous styrene–dioxane solution gave the highest stabilization for a given amount of retained polymer.

Foreign Literature on Radiation-Induced WPC

A review of the foreign literature on radiation-induced WPC, like one of the domestic literature, is more than ordinarily complicated by choice of scope. If the effects of radiation are ignored, the reviewer has an easier task. These effects could have a practical bearing on the problem of producing satisfactory WPC by irradiation since, at doses of 5 to 10 Mrads, there is some evidence of structural damage. However, Kent notes* that radiation doses ordinarily used or contemplated for commercial processing (0.5 to 3 Mrads) have negligible effect on wood properties.

Only part of the dilemma is faced in this review. This section discusses foreign literature on radiation-induced WPC and recent studies on the effects of radiation on wood. An approach to coping with the overwhelming amount of literature on radiation effects on wood constituents (e.g., cellulose, lignin) and WPC materials (e.g., vinyl chloride, styrene) may be provided by current Oak Ridge National Laboratory–*Nuclear Science Abstracts* attempts to computerize the abstract literature in nuclear science, including that on radiation polymerization. Hopefully, the scope of this work will be broad enough to encompass radiation effects on wood as well as WPC materials to permit complete evaluation of radiation effects.

However, no review of the literature has appeared that comprehensively encompasses

*Personal communication, Oct. 6, 1965.

both the effects of radiation on wood and the radiation polymerization of WPC materials, even at low doses. In addition, as Siau et al.[24] have pointed out, the small effects of radiation on cellulose should not be completely ignored in discussing WPC.

An indication of recent USSR interest in WPC is apparent from two articles appearing in 1964 by Kalnin'sh, of the Latvian Academy of Sciences. One of these is a popular review[54] of "plastification" of wood, and the other[55] mentions the radiation-induced polymerization of formaldehyde, acetone, and other compounds in wood. Kalnin'sh also gives data on the solubility of wood in benzyl alcohol and water after treatment with radiation and ammonia, indicating that research in basic mechanisms of radiation effects on wood still is being actively pursued. Freĭdin, Malinskiĭ, and Karpov,[56] who pioneered this work in the Soviet Union, described the effects of gamma radiation on wood properties and composition at a conference in Tashkent in 1961. Dose rates of 0.6 to 0.9 Mrad/hr were studied. These workers[13] apparently were among the first to report information concerning the effect of irradiation of wood both in the natural and in the treated state. The effect of gamma radiation on wood at doses of 1.5×10^9 rads was reported recently by Bonotto et al.[57] at Mol, Belgium. They found a decrease in volume, density, and weight, a large increase in simple sugar content, and the appearance of arabinose and xylose in irradiated wood. Depolymerization was explained by direct and indirect action of gamma radiation through formation of peroxides from the water in the wood.

The chemistry of irradiated wood has been extensively investigated in Germany. Seifert[58] notes that, although high-intensity radiation causes a reduction in the mechanical strength of wood, low-dose irradiation increases the strength. Tests were made with 10-mm-thick boards (25 × 5 cm) of pine, birch, and spruce irradiated with a ^{60}Co source from 10 to 10^8 r at 40°C. Before and after irradiation the wood samples were analyzed for cellulose, lignin, pentosans, carboxyl and methoxyl groups, alkali solubility, sugars, alcohol extracts, chain length, and fine structure. No new discoveries were made in the high-dose range (10^4 to 10^8 r), but in the range from 10 to 10^3 r, high polymers of wood disintegrated. This loss was more than compensated for by the partial cross-linking of the wood constituents. Conclusions based on chromatographic and roentgenographic observations indicated that gamma rays first decomposed some parts of the wood micelles, which were still in their lattice-like order, and acquired an even higher degree of order through intermolecular connection. The new degree of order, it was concluded, might be the cause of the increased strength at these low doses. In the work of Lueck and Dell,[59] doses above 1 Mrad caused a marked degeneration of cellulose, especially a splitting of the 1,4-β-glucosyl bond. Numerous breakdown products appeared, such as mono-, di-, and oligosaccharides, cellulodextrins, acids (2-ketogluconic acid and its lactone, 2-ketocellobionic acid), reductones, and gases ($CO > CO_2 > H_2$). With high radiation doses, carboxylated cellulose was not polymerized, but the methyl, hydroxyethyl, and methylhydroxybutyl derivatives were. At higher doses, oxidations and splitting off of the substituents occurred. Polymerization reactions were favored by a low viscosity of the medium, whereas degradation reactions predominated in a high-viscosity medium. Lignin, which has a protective effect on cellulose, was less sensitive to irradiation than cellulose, an effect noted by Karpov et al.[13] Hemicelluloses were much easier to degrade than cellulose. Pectins too, especially protopectins, were very sensitive to ionizing radiation at doses over 0.1 Mrad. On the other hand, pectin jellies made from sugar, acid, and pectin were affected only by doses above 1 Mrad.

Australian workers[60] investigating the effects of radiation on eucalyptus wood meal found that, up to a dose of 10 Mrads, radiation had only a slight effect; the ethanol solubility of the wood increased from 0.4 to 1.5%, and the solubility in 0.5% aqueous NaOH increased from 9.8 to 18.4%. Above 10 Mrads the effect was considerable, and at about 300 Mrads the ethanol solubility of the wood was 30%, from which 8.2% lignin with 23.1% hydroxymethyl radical was recovered; the alkali solubility was 57.4%, from which 8.8% crude lignin was obtained.

Japanese workers, who have been extremely active in radiation polymerization and grafting research, have also studied extensively the effects of radiation on cellulose.[61] Hoin[62] recently compiled a literature survey on radiation-induced graft polymerization on cellulose. He cites 94 references to the report and published

literature; major sources were *Nuclear Science Abstracts, Chemical Abstracts,* and *Chemical Titles.* The years covered are 1959 to 1964.

According to Youngs,[63] research at the British Forest Products Research Laboratory indicated that 10 to 60% content (dry weight of wood basis) of methyl methacrylate could be polymerized in scotch pine sapwood with a dose of 1.5×10^7 rads. The British workers observed little penetration of the monomer in the cell wall, and the results were not satisfactory. It should be noted that other workers have obtained satisfactory WPC without penetration of monomer into the cell wall (see Fig. III-5). Youngs also discusses some of the American Society for Testing and Materials testing procedures and evaluation of wood properties by nuclear techniques. (D. A. Fuccillo)

Technical and Economic Considerations for an Irradiated Wood–Plastic Material

An evaluation of the commercial utility of irradiated wood–plastic combinations (WPC) indicated that these materials should find their widest application in products where raw-material cost is small relative to production cost, where the esthetic qualities of wood are important, or where those enhanced physical properties which the process imparts to wood are of special value. The potential applications include fabricated materials of construction (stair treads, counter tops, and flooring), furniture, sporting goods, and specialty items. The estimated dollar costs for 2500-, 1250-, and 625-lb/hr processing plants are $1,560,000, $1,045,000, and $743,000 for investment capital and $676,000, $478,000, and $391,000 annual operating cost, respectively. Recommendations are made for further establishing the utility of the product. This evaluation was made by Arthur D. Little, Inc.

Arthur D. Little, Inc., at the request of the DID, AEC, completed a study[64] to determine the commercial utility of irradiated wood–plastic combinations (WPC). Both technical and economic aspects were considered, with data developed by West Virginia University and Air Reduction Company, Incorporated, and evaluations by Arthur D. Little, Inc., being used to characterize the various products that could be made by this method. Costs were estimated on the basis of variations in monomer type, dosage, source efficiency, and plant capacity, with vacuum impregnation assumed in all cases.

About 38 billion board feet of wood are used annually in this country; of this market, 4.9 billion feet are used in applications where the cost of the wood is low relative to the overall product cost. It is in this usage and where the esthetic qualities are important that WPC should find their greatest application. The fraction of this specialty market that the new product will capture will depend on cost–performance characteristics.

Some conventional methods of wood treatment impart desirable properties to wood, and in many instances WPC combine the advantages of several of these treatments. Conversely, some treatments give characteristics not available in WPC. Impregnation with preservative oils and salts (zinc chloride, arsenites, acid copper chromate, etc.) provides good decay resistance but does little to enhance other physical properties. Impregnation with a thermosetting resin improves dimensional stability, hardness, compression strength, and decay and rot resistance; however, phenolic resins discolor the wood and lower the tensile and impact strengths, while urea resins hydrolyze at elevated temperatures and high humidities. Heat and chemical treatments, although providing in most cases some dimensional stabilization, frequently degrade the physical properties of the wood. Thus it appears that, although conventional treatments impart certain desirable properties to wood, no single treatment possesses all of the advantages.

Manufacturing Process

Wood is vacuum impregnated with a suitable monomer and then irradiated to polymerize the monomer within the wood (Fig. III-12). Since oxygen inhibits the polymerization of some monomers, e.g., vinyl acetate (Fig. III-13) and methyl methacrylate, woods impregnated with these are irradiated in the absence of oxygen. Removal of dissolved oxygen from these monomers shortens the irradiation induction period and reduces the dosage requirements. Addition of an activator (e.g., carbon tetrachloride) also reduces the dosage required by methyl methacrylate. Polymerization of vinyl chloride is not inhibited by oxygen, but vinyl chloride is a gas under ordinary conditions, and when it is used, 3 atm pressure is re-

quired during the irradiation to keep it in the liquid phase and in the wood.*

VACUUM IMPREGNATION

Vacuum impregnation, although more expensive than conventional dip or spray application, has the advantages that irregular shapes are easily treated and that the depth of penetration can be controlled by regulating the immersion time and pressure. Impregnation conditions must be evaluated for each specific end use of the product.

Vacuum impregnation is a batch process. In a typical cycle with oxygen-sensitive monomers, the container holding the wood is evacuated to open the pores and remove the oxygen, nitrogen is admitted, the container is reevacuated, and then the monomer is introduced under 1 atm nitrogen pressure and covers the wood. The minimum cycle time is about 3 hr for a single nitrogen purge; a second nitrogen purge to further reduce the amount of oxygen would require an additional hour. The treated wood is removed from the impregnation chamber and encapsulated to protect the monomer from oxygen during the subsequent irradiation. Polyethylene appears useful for protection since it provides an adequate barrier, is inexpensive, and will not reduce the irradiation efficiency.

IRRADIATION

The choice between isotope and machine irradiators is dictated by the dose rate and shape of the material to be irradiated. Isotopic irradiators, ^{60}Co for example, are used for in-depth treatment or for irregular objects, and machine irradiators are normally used for flat samples.

Radiation polymerization is not new, and, according to one authority,[66] few if any economic advantages are to be gained by sub-

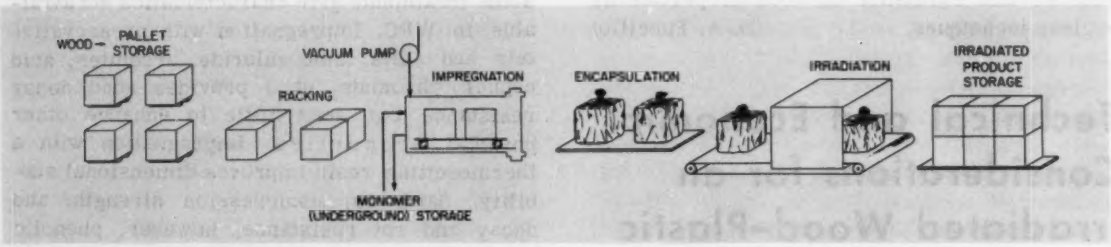


Fig. III-12 Flow diagram of manufacturing process for irradiated WPC.

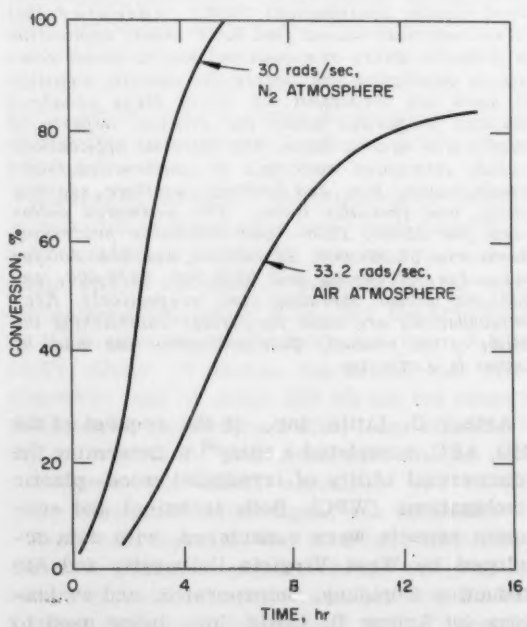


Fig. III-13 Effect of inert atmosphere on polymerization rate of vinyl acetate in birch sapwood. (Data developed by West Virginia University.)

*More recent work[65] by Air Reduction Company, Incorporated, on polyvinyl chloride-impregnated WPC indicates that the use of pressurized containers for the volatile vinyl chloride can be avoided by irradiating at a temperature of −78 °C, maintained with dry ice. At this temperature vinyl chloride is a liquid (vapor pressure, 16 mm) and also forms a polymer of higher molecular weight than that formed at higher temperatures. The method was satisfactory for a variety of woods, and samples with density increases up to 100% were produced. Some wood–polyethylene combinations were also prepared and showed improvement in dimensional stability, the only property tested.

stituting radiation for a conventional free-radical catalyst. However, economic and technical advantages may accrue in the radiation-induced WPC manufacture in those cases where (1) monomer pot life is extended in the absence of catalyst; (2) controlled polymerization prevents heat buildup in thick sections; (3) cold curing eliminates heating and accompanying monomer evaporation problems; and (4) no catalyst residue remains in the wood.

The irradiation process is simple to control, and the upper and lower dose limits are not critical with WPC. The irradiation time can easily be changed merely by changing the speed of the conveyor belt through the irradiation chamber. Unlike some commercial processes, heat and pressure are not required in the radiation process.

PROCESS VARIABLES

The variables that have the most effect on the properties of WPC are wood species, type of monomer, distribution of monomer within the wood, and additives.

Wood Species. The uniformity, depth of penetration, types of monomers that can be used, and, to a large extent, the ultimate characteristics of the product are determined by the wood species. The compatibilities of various WPC are listed in Table III-6. Air Reduction Company, Incorporated, recently prepared samples of the following woods with various amounts of polyvinyl chloride:

Ponderosa pine	Red oak
Red cedar	Yellow pine
Poplar	White fir
Douglas fir	Philippine mahogany
Walnut	Basswood
African mahogany	Redwood
Birch	

Table III-6 COMPATIBILITY OF WOODS WITH PLASTICS*
Irradiation: 3.2×10^6 rads (10^4 rads/hr)

Wood	Compatibility†			
	Methyl methacrylate	Vinyl acetate	Ethylhexyl acrylate	Ethyl acrylate
White pine	G	Q‡	Q§	Q§
Spruce	G	Q‡	Q	Q
White oak	G	G	Q	Q
Red oak	G	G	Q	Q
Yellow poplar	G	G	Q	Q
Sugar maple	G	G	Q	Q
Beech	G	G	Q	Q
Birch	G	G	Q	Q
Locust	G	G	Q	Q
Mahogany	G	G	Q	Q
Redwood	Q‡	Q‡	Q	Q

*Data developed by West Virginia University in exploratory tests.

†G = apparently good; Q = questionable.

‡Irradiated samples contained unreacted monomer after a total dose of 6.4 Mrads (1×10^4 rads/hr).

§Both ethyl acrylate and ethylhexyl acrylate appeared to polymerize completely but caused severe splitting of the wood. The same effect was observed earlier with sugar maple and with butyl acrylate.

Type of Monomer. Much of the work has been done with methyl methacrylate (MMA), styrene, vinyl acetate (VA), and vinyl chloride (VC) because of their cost and comparative ease of radiation polymerization. Monomers that have been evaluated in combination with sugar maple are:

Methyl methacrylate	Tetrahydrofurfuryl methacrylate
Vinyl chloride	1,3-Butylene methacrylate
Styrene	Triethylene glycol dimethacrylate
Cyclohexyl methacrylate	Tetraethylene glycol dimethacrylate
Allyl methacrylate	Butyl acrylate
Hydroxyethyl methacrylate	Methyl acrylate
Ethylene dimethacrylate	Ethyl acrylate
Diaminoethyl methacrylate	2-Ethylhexyl acrylate
t-Butylaminoethyl methacrylate	

Distribution of Monomer. In vacuum impregnation of wood, a large concentration gradient exists between longitudinal and transverse directions of the wood; i.e., plastic penetrates into the end of the grain at a faster rate than into the surface under a given set of conditions. Figure III-14 shows the gradient in transverse density of birch and pine impregnated with methyl methacrylate. Wood species, wood section (i.e., heart vs. sapwood), immersion time, size of the sample, and viscosity of the monomer all affect the monomer distribution and impregnation cycle. In practice, the impregnation cycle conditions should be optimized for each specific item.

Additives. Some materials considered as additives to the monomer to further enhance desirable characteristics of the wood were fungicides, stains and dyes, bacteriostats, ultraviolet absorbers, flame retardants, and odorants. West Virginia University has already undertaken an evaluation of some of these additives, but much work remains to be done.

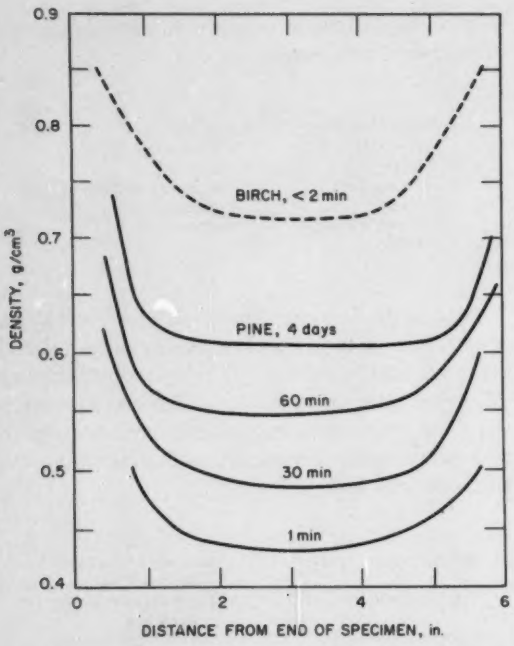


Fig. III-14 Gradient of transverse density in birch pretreated at 300 mm Hg abs. and in white pine pretreated at 400 mm Hg abs. for 20 min, followed by immersion in methyl methacrylate for different times. Samples: sapwood, 2 by 2 by 6 in. (Data from West Virginia University.)

Properties of WPC

The texture and appearance of natural wood are retained in WPC, and the esthetic qualities are one of the principal advantages of these materials. Finishing, refinishing, and maintenance should be easier with the polymer in the wood, and repairing the finish might be possible by burnishing and melting the thermoplastic polymer to form a continuous surface.*

Most of the properties of WPC are superior to those of untreated wood, with the possible exception of nailability and impact resistance.† Hardness, static bending, shear strength, abrasion resistance, flame retardancy, and water absorption have been tested, and tests on stain resistance, impact strength, and weatherability were under way at the time of writing of the report. Standard or slightly modified ASTM tests are used.

Wood–plastic combinations are harder than untreated wood, the hardness increasing with increasing polymer content (Fig. III-15). Both static bending and shear strength increase slightly with increased polymer content (Fig. III-16). However, the increase is not significant and would not justify the increased cost. All samples tested except red cedar (the exception

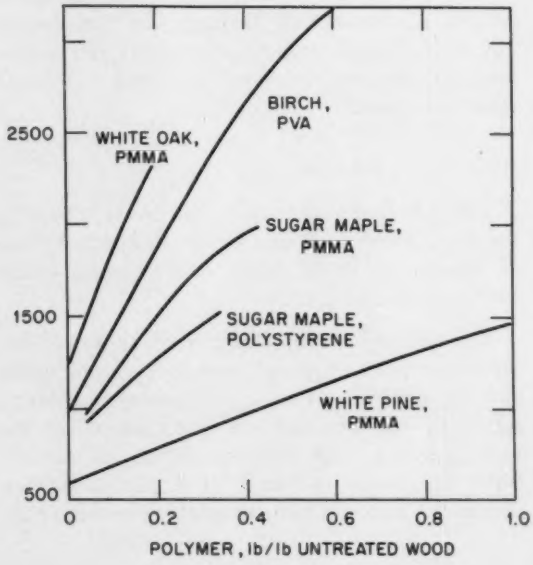


Fig. III-15 Effect of polymer content on hardness of WPC.

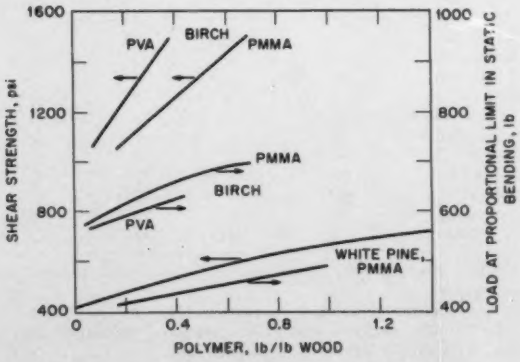


Fig. III-16 Effect of polymer content on static bending and shear strength of WPC.

*Recent information shows that it is possible to restore the finish by this method on WPC damaged by scratches, cigarette burns, etc. (Private communication from M. H. Stein.)

†More recent information from Kent, University of West Virginia, indicates the impact resistance of WPC to be superior to that of untreated wood. (Private communication.)

Table III-7 EFFECT OF POLYMER (PVC) ON ABRASION RESISTANCE OF WPC*

Wood	PVC, lb/lb wood	Vol. loss, %		Improvement, %†	
		Fine abrasive	Coarse abrasive	Fine abrasive	Coarse abrasive
Ponderosa pine	0.09	0.19	0.98	42	8
	0	0.27	1.07		
Red cedar	0.24	0.88	0.83	−31	10
	0	0.67	0.91		
Poplar	0.48	0.36	0.45	55	21
	0	0.56	0.57		
Philippine mahogany	0.62	0.19	0.55	100	100
	0	0.39	1.14		
Basswood	0.81	0.25	0.55	52	420
	0	0.38	2.89		
Redwood	1.0	0.44	1.62	160	0
	0	1.16	1.56		

*Data developed by Air Reduction Company, Incorporated.
†The % improvement = 100(untreated − treated)/treated.

may be an experimental error) showed a significant improvement in abrasion resistance (Table III-7), which is one of the most important characteristics in several potential applications. Only vinyl chloride test data are available, but tests with other monomers are in progress. Flame retardancy appears to be better than for untreated wood (Table III-8). Additional tests using various levels of monomer and other wood species are in progress.

Table III-8 EFFECT OF POLYMER (PVC) ON COMBUSTIBILITY OF RED CEDAR WPC*

PVC, lb/lb wood	Weight loss in burning, %	Time, sec	
		To cease burning	To cease glowing
0.24	83.3	318	1035
0	91.4	450	875

*Data developed by Air Reduction Company, Incorporated. For recent additional data see Ref. 65.

Water absorption was lower in several of the treated woods, the rate being based on the percentage of water absorbed as calculated both for wood alone and for wood plus plastic (Table III-9). The dimensional stability of wood varies with moisture content. This instability, which has long been recognized as a limiting property of wood, is decreased as a result of

Table III-9 EFFECT OF POLYMER (PVC) ON WATER ABSORPTION OF WPC*

Wood	PVC in treated wood, lb/lb wood	Weight increase after 24-hr immersion, %		
		Treated wood		
		Combination basis†	Wood basis‡	Untreated wood§
Philippine mahogany	0.78	7	12	36
Walnut	0.19	17	20	26
African mahogany	0.04	33	34	31
Birch	0.32	10	13	19
Red oak	0.24	11	14	54
N. C. yellow pine	1.00	17	34	124
Redwood	1.34	5	11	29
Hemlock	0.76	20	35	45
White fir	1.37	25	59	34
Douglas fir	0.54	11	17	19
Western red cedar	0.26	26	33	29

*Data developed by Air Reduction Company, Incorporated.

†The % increase = $\frac{\text{Weight of absorbed water}}{\text{Weight of combination}} \times 100$.

‡The % increase = $\frac{\text{Weight of absorbed water}}{(\text{Weight of combination}) - (\text{Weight of polymer})} \times 100$.

§The % increase = $\frac{\text{Weight of absorbed water}}{\text{Weight of untreated wood}} \times 100$.

the lower water-absorption rates, and thus variations in humidity will have less effect on treated wood used for flooring, fabricated construction parts, outside seating, and sporting goods. Weatherability tests indicated WPC to be superior to untreated wood and about equal to wood that is conventionally finished. Further tests are being made to verify these findings and evaluate more promising samples. Forest Products Laboratory reports that fungus and rot resistance are improved. This probably results from lower porosity of the product which should enhance its general chemical properties.

Although no quantitative test to define nailability was used, attempts to drive six-penny finishing nails through samples of white pine containing 35% vinyl acetate indicated a sharp reduction in this quality (resistance to splitting when nailed), and splitting was a serious problem.

West Virginia University personnel have observed that fabrication operations—turning, tapping, planing, and wood finishing—are easier with WPC than with either wood or polymer alone.

Economics of WPC

Before commercial uses of this new product can be considered, its cost must be estimated. In addition to characterizing the physical properties, Arthur D. Little, Inc., made an order-of-magnitude estimate of manufacturing costs for various WPC. An independent service facility was considered, and total investments for land, buildings, and operating facilities were included. Such a facility could, of course, be an integral part of an existing wood-processing plant; then the total outlay for these items would be considerably less, and manufacturing costs would decrease considerably.

The total manufacturing cost was broken into three components: impregnation, irradiation, and raw materials. Plant operating and investment costs were developed separately for the first two items to better identify the various increments of total operating cost. Wood–plastic combinations production rates of 2500, 1250, and 625 lb/hr (20, 10, and 5 million lb/year) were selected to illustrate the sensitivity of operating cost to plant capacity, and economics were developed separately for each. These production rates correspond to ^{60}Co sources of 1 million, 500 thousand, and 250 thousand curies, respectively, and are based on a 1-Mrad dose requirement, 25% source utilization, and 90% mechanical efficiency of the plant. Vacuum impregnation and irradiation investment costs were developed for the 2500-lb/hr plant, and appropriate factors were used to estimate the investment for the smaller plants. The costs are summarized in Table III-10.

Table III-10 SUMMARY OF ESTIMATED INVESTMENT AND OPERATING COSTS FOR WPC PRODUCTION

Production, lb			Initial investment, dollars	Operating cost	
Per hour	Per year			Annual,* dollars	Unit, cents per pound of product
2500	20×10^6	Impregnation	280,000	224,000	1.12
		Irradiation	1,280,000	452,000	2.26
		Total	1,560,000	676,000	3.38
1250	10×10^6	Impregnation	190,000	174,000	1.74
		Irradiation	855,000	304,000	3.04
		Total	1,045,000	478,000	4.78
625	5×10^6	Impregnation	130,000	150,000	3.00
		Irradiation	613,000	241,000	4.82
		Total	743,000	391,000	7.82

*See footnote on page 137.

VACUUM IMPREGNATION COST

Investment costs for vacuum impregnation are based on a batch cycle established for a particular wood species, monomer, sample size, etc.; a one-batch cycle of 8 hr is assumed for vinyl acetate and methyl methacrylate. This period includes time for loading and unloading the impregnator, evacuating the chamber, double-purging it with nitrogen, introducing the monomer into the impregnator, and allowing the desired amount of monomer to soak into the wood. The investment cost of $280,000 and the annual operating cost of $224,000 for the 2500-lb/hr impregnation facility are broken down in Table III-11. The annual operating cost of $224,000 corresponds to about 1.1 cents per pound of product, excluding the monomer cost. The impregnation cost decreases with increasing production rate (Fig. III-17).

IRRADIATION COSTS

Cost information developed by Brookhaven National Laboratory[67] and Arthur D. Little,

Table III-11 ESTIMATED PLANT INVESTMENT AND OPERATING COSTS FOR VACUUM IMPREGNATION OF WOOD WITH MONOMER

Basis: 2500-lb/hr (20×10^6 lb/year) Production, 8-hr Batch Cycle, 8000-hr/year Operation

	Cost	
Major process equipment, installed		
Impregnator, 6- by 50-ft cylinder (includes vacuum pump, drive, explosion-proof motor)	$75,000	
Monomer storage tanks, lined, 15,000 gal (2 included)	30,000	
Surge tanks, lined, 5000 gal (2 included)	14,000	
Vacuum packing machine	10,000	
Materials-handling equipment (forklift truck, $7000; impregnator trolleys, $2000; racks for dimensioned parts, $8000)	17,000	
Total installed-equipment cost (IEC)		$146,000
Plant equipment		
Electrical, 5% IEC	8,000	
Piping, 20% IEC	29,000	
Fire-protection equipment	10,000	
Total plant-equipment cost (PEC)		193,000
Physical plant		
Land and site preparation (1 acre)	5,000	
Building, storage, and factory control laboratory (5000 sq ft at $8.00 per square foot)	40,000	
Total physical-plant cost (PPC)		238,000
Fixed capital		
Engineering and construction, 12% PPC	28,000	
Contingency, 6% PPC	14,000	
Total fixed capital (FC) (excluding working capital)		$280,000
Annual manufacturing cost (excluding monomer)		
Depreciation, 10% FC per year	28,000	
Direct labor	60,000	
Factory overhead	60,000	
Maintenance labor and supplies, 3% FC	8,500	
Utilities, 2% FC	5,500	
Local taxes and insurance, 3% FC	8,500	
Operating supplies, 3% FC (including N_2 and polyethylene film)	8,500	
Total manufacturing cost		$179,000
General expenses, 25% manufacturing cost	45,000	
Total operating cost		$224,000

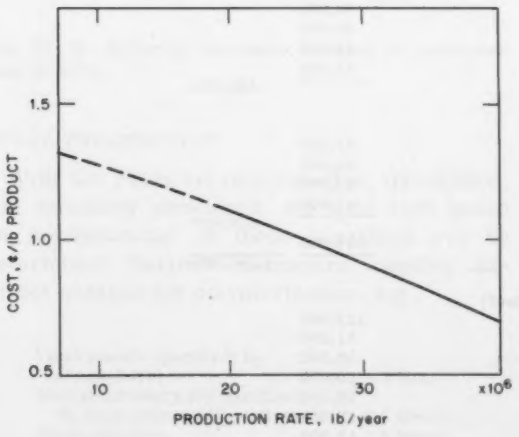


Fig. III-17 Effect of production rate on cost of vacuum impregnation. Nominal production rate 2500 lb/hr, investment $280,000, nominal operating cost $224,000 per year.

Inc.,[68] was used in estimates of plant investments and process economics.

The nuclide cost may be treated as fixed capital to be depreciated over the life of the plant or as an annual expense. In the cost estimates for the three sizes of plants, it was considered as part of the plant investment to be amortized over the life of the equipment.

The cost of cobalt with an activity of 30 curies/g is based on an AEC price of 50 cents per curie* plus 15 cents per curie* cost for en-

*On Dec. 18, 1965, AEC announced a price reduction for ^{60}Co with specific activities from 1 to 15 curies per gram; the price is $0.40 per curie for 100,000 or more curies per order. All ^{60}Co costs included in this review (and the Arthur D. Little report) are subject to this price change providing the lower activity ^{60}Co can be conveniently used. In addition, current encapsulation costs must also be considered.

capsulation and transportation. Fixed capital for a 1-Mc source (for a 2500-lb/hr plant) is therefore $650,000. The irradiation facility cost, less the cobalt, is estimated to be $630,000, giving a total of $1,280,000 fixed capital. A 10-year straight-line depreciation plus a source replacement allowance was used, with the assumption that the venture would be financed from equity funds, thus excluding interest on borrowed capital. A breakdown of costs for the irradiation facility is given in Table III-12.

The irradiation operation cost will be a function of dosage requirements and source efficiency. Dose requirements for polymerization will vary from 0.5 Mrad for vinyl acetate to 1.5 Mrads for methyl methacrylate, and source utilization can vary from 15 to 30% with variations in product geometry. Unit cost may also be expressed by use of the production rate regardless of whether its variations result from variations in either dosage requirements or source efficiency. On this basis, a basic production cost of 1.9 cents per pound is esti-

Table III-12 ESTIMATED PLANT INVESTMENT AND OPERATING COSTS FOR IRRADIATION OF WPC

Basis: Production, 2500 lb/hr, 20×10^6 lb/year; Operation, 8000 hr/year; Rated Activity, 1 Mc (^{60}Co) At $0.65 Per Curie

	Cost	
Major process equipment, installed		
Pool	$ 20,000	
Source-handling tools	5,000	
Pool deionizing system	10,000	
Source elevator and product conveyor	60,000	
Source-holding frames	5,000	
Instrumentation	20,000	
Materials-handling equipment	7,000	
Total installed equipment (IEC)		$ 127,000
Plant equipment		
Electrical (10% IEC)	13,000	
Piping (10% IEC)	13,000	
Laboratory equipment	20,000	
Painting, ventilation	10,000	
Outside lines (electrical and water)	5,000	
Total plant and equipment cost		188,000
Physical plant		
Land and site preparation	30,000	
Buildings		
Foundations	40,000	
Irradiation cell	80,000	
Production and office area	30,000	
Mechanical service	25,000	
Electrical service	35,000	
Total physical-plant cost (PPC)		428,000
Fixed capital		
Design	80,000	
Licensing, startup, training	36,000	
Construction (10% PPC)	43,000	
Contingency (10% PPC)	43,000	
Subtotal fixed capital		630,000
^{60}Co source: 1 Mc at $0.65 per curie	650,000	
Total fixed capital (FC)		1,280,000
Annual manufacturing cost (excluding monomer)		
Depreciation, 10% per year	128,000	
Replacement of source, 12.5% per year	81,000	
Direct labor	50,000	
Factory overhead	50,000	
Technical	10,000	
Maintenance and utilities, 3% FC	19,000	
Local taxes and insurance, 3% FC	19,000	
Operating supplies, 1% FC	6,000	
Total manufacturing cost		363,000
General expenses, 25% manufacturing cost	89,000	
Total operating cost		$ 452,000

mated for the 1-Mc source, assuming a 25% source efficiency and a 1-Mrad dose.

MONOMER COST

The monomer as a component of the manufacturing cost was considered separately since the amount and type affect the ultimate physical properties of the product. Figure III-18 illustrates the cost of three monomers at various loading levels.

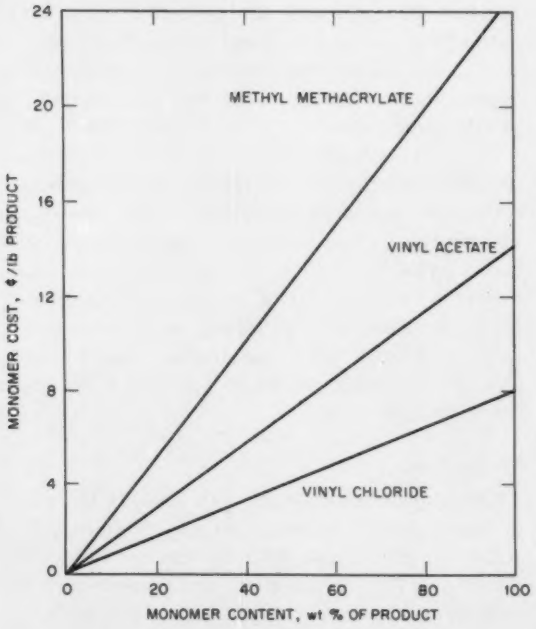


Fig. III-18 Effect of monomer content on monomer cost of WPC.

TOTAL PROCESS COST

With the costs for impregnation, irradiation, and monomer developed, the total cost based on combinations of these variables can be determined. Various monomers require different dosages for polymerization, e.g.,

Vinyl acetate (purified; N_2 atmosphere)	0.5 Mrad
Methyl methacrylate (purified; N_2 atmosphere; CCl_4 activator)	1.5 Mrads
Vinyl chloride	0.6 Mrad

The specific dosage requirement would be affected by the dose rate, the species of wood, and the monomer used. The above values are average dosage requirements at a dose rate of about 30 rads/sec. If lower total doses are required, of course the plant throughput is higher. For example, a plant based on 1-Mrad dosage for 20 $\times$ 10^6-lb/year production of material requiring 1 Mrad of irradiation for polymerization could produce 33 $\times$ 10^6 lb/year of a product with vinyl chloride monomer (0.6 Mrad required dosage) but only 13.3 $\times$ 10^6 lb/year of a product with methyl methacrylate (1.5 Mrad required dosage). Figure III-19 shows the relation between production

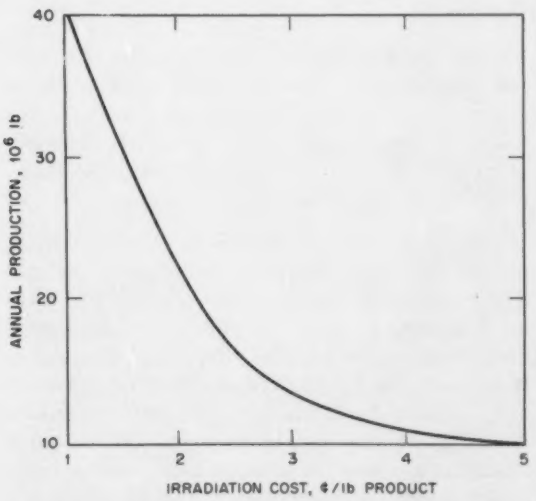


Fig. III-19 Effect of production rate on cost of irradiation. 1-Mc source; annual operating cost $452,000; 8000 hr per year of operation; 90% mechanical efficiency.

rate and irradiation cost. For example, for applications requiring 25% polymer and production adjusted as described above for a fixed source, production costs would be:

Process variable	Cents per pound of product: Vinyl chloride	Methyl methacrylate
Vacuum impregnation	0.9	1.2
Irradiation	1.4	3.1
Monomer	2.0	6.0
Total basic cost	4.3	10.3

Obviously, the monomer is the largest single production expense.

Utility of WPC

In screening possible applications for WPC, the principal requirements for each end use must be identified. Information was supplied by manufacturers, industrial associations, and the Arthur D. Little, Inc., staff. The assumption was made that properties of WPC can be tailored for a specific application. Lists of potential applications were prepared, and each application was evaluated according to requirements and performance. Six major categories of applications were considered.

CONSTRUCTION

Cost considerations quickly ruled out WPC for high-volume low-unit-cost uses such as framing, sheathing, siding, and shingles. Remaining are the high-unit-cost items: flooring, fabricated units, e.g., doors and windows, laminated wood, and paneling.

Flooring. The properties of WPC correlate nicely with the abrasion resistance, permanence of finish, appearance, resilience, ease of fabrication, and low-moisture-absorption rate required for specialty flooring. The use of this material for basketball flooring presumably would facilitate or possibly eliminate finishing at the installation site. Its abrasion resistance, ease of maintenance, and permanence of finish might justify the 20 cents per square foot (7 cents per pound) increase in cost over that of some conventional flooring. Requirements for bowling-lane flooring are about the same as those for basketball flooring, with the additional requirement of impact resistance. Wood–plastic combinations might also be used for the tail planks, kickback plates, and gutters. Maple–plastic combinations might find use in parquet flooring, although they are more expensive than the polyurethane-coated materials now used.

Fabricated Units. Wood–plastic combinations should find application for window sashes and frames and outside doors if they prove to be durable and have good weatherability, permanence of finish, and decay and rot resistance. The treatment appears to be less expensive than the materials and labor required to paint and finish both the interior and exterior of window frames at the installation site. The abrasion resistance, in-depth finish, appearance, permanence of finish, and mar resistance of the product should also be useful in stair treads, handrails, and kitchen-cabinet doors and counter tops.

FURNITURE

Wood–plastic combinations have the properties, appearance, mar resistance, stain and water resistance, and hardness and permanence of finish required for veneer for table, desk, bench, and counter tops. At an estimated 29 cents per square foot (which includes a 25% return in investment), WPC should be competitive in cost with the high-pressure laminates that retail for about 30 cents per square foot and frequently attempt to simulate the appearance of wood. That they can compete in performance remains to be demonstrated.

In solid furniture for churches and auditorium benches and chairs, the added cost of the treatment that could be tolerated would depend on the degree of improvement that the treatment could provide. Outdoor stadium seating, which requires weatherability, decay and rot resistance, permanence of finish, mar resistance, ease of fabrication, resistance to splintering, and dimensional stability, appears to be a potential application.

INDUSTRIAL USES

The high cost of WPC may exclude their use in many industrial applications. They may find utility in electrical and high-energy fields, but these are less promising than those where the esthetic qualities of wood are important.

SPORTING GOODS

Because of their appearance, weatherability, impact strength, permanence of finish, and high strength-to-weight ratio, WPC find some application in sporting goods. If the impact strength of treated-wood heads of golf clubs is sufficiently high, the conventional vulcanized fiber insert might be eliminated. Its attachment to the club head is an expensive operation, and additional cost equal to that of the fabrication can be tolerated. The insert is moisture sensitive, and professional golfers claim that its dimensional instability causes "off-angle performance."

Gunstocks for sporting rifles appear to be another promising application for the product. If the resin facilitates the finishing as anticipated and improves other physical characteristics, an additional cost of $1.50 to $2.00 over

the approximate $3.50 cost for a conventional walnut gunstock blank would not be unreasonable. The additional weight, depending on the resin concentration required for a satisfactory finish, could be compensated for by using a lower density wood. The splintering resistance and durability must be as good as that of walnut.

In the boating industry, composites probably could find use only in trim applications where appearance is important, since the use of wood as a structural material is rapidly declining in boat manufacturing.

TOYS AND SCHOOL SUPPLIES

School desk tops, chairs, and workbenches are the most promising applications in this category. Ease of maintenance and repair, which is related to permanence of finish, helps the product meet the more rugged demands of school furniture. The flame-retardance property, although a desirable feature, would not command a much higher dollar price.

The use of WPC for toys looks unlikely because of cost.

SPECIALTIES

The most promising applications of WPC for specialty items are cutlery handles, wooden ware (e.g., salad and fruit bowls), and cutting boards. Appearance, permanence of finish, ease of fabrication, and stain resistance are the attractive characteristics. The conventional high unit cost of these items makes the use of combinations appear practical; however, they must demonstrate satisfactory performance when exposed to repeated washings.

Market Volume

The principal objective of this study was to identify the commercial utility of WPC rather than to make a more conventional market study. However, an attempt was made to develop order-of-magnitude estimates for the size of market and the add-on value that could be anticipated for WPC even though costs and properties essential for an evaluation were not fully established. Manufacturers had no product for comparison with those they were using.

Estimates for markets in which WPC would compete were provided by a summary of lumber consumption in the potential applications (Table III-13). The total, 4.9 billion board feet, is not the estimate of the lumber market that WPC can hope to capture, but rather an overall lumber consumption in the applications described. At an estimated 20 to 40 cents per board-foot purchase price to manufacturers, this market represents an annual $1 billion to $2 billion industry. A 20 to 40 cents per board-foot purchase price to a manufacturer seems a reasonable range for the wide price variation encountered in the suggested end-use applications. A market volume, estimated on the raw material going into manufacture of converted products, can more readily be identified as the dollar volume of the lumber component than as the total sales volume of the industry, which may include a substantial portion of nonlumber raw materials. With an additional $200 million market for plastic laminates, the total market in which WPC would be competing may be crudely estimated at $1.2 billion to $2.2 billion annually.

Table III-13 ESTIMATED LUMBER CONSUMPTION IN POTENTIAL APPLICATIONS OF WPC (1960)

Applications	Million board feet
Construction	
Specialty flooring	35
Fabricated units (window sashes and dividers, doors, stair treads, handrails, cabinet doors)	2200
Furniture	
Commercial and public	107
Household	2088
Industrial	
Textile (bobbins and shuttles)	26
Electrical	70
Patterns and jigs	71
Sporting goods	
Golf-club heads, gunstocks, boats, bowling pins, squash and badminton rackets	137
Toys and school supplies	
Toys and games	56
School supplies (other than desk tops and chairs) included with commercial and public furniture	
Specialties	
Hand and garden tools	88
Cutlery handles, wooden ware, cutting boards, slide rules	5
	4883

Recommendations

It is the opinion of Arthur D. Little, Inc., that the overall development program for WPC should be two-phased and include both commercial development and applied research.

The commercial development program would include a systematic study to supplement existing information on various properties as a function of monomer type, monomer loading, wood species, and wood quality. Samples for end-use testing would be prepared and compared directly with materials now used. Minor variations in existing WPC manufacture, such as addition of wood stains and dyes, ultraviolet absorbers, and preservatives, would be included. Processing variables would be investigated to define more clearly manufacturing procedure and operating cost as an integral part of preparation of samples for end-use testing. Samples of possible products should be solicited from manufacturers, treated at the estimated acceptable level with the most promising monomer, and submitted to the potential manufacturer for testing. Field trips revealed a willingness by possible investors to participate in such an evaluation program.

The applied research program would be concerned with achieving order-of-magnitude cost reduction and product improvement. Cost-reduction studies should include investigations of various monomers and processing techniques, which would permit use of lower unit-cost monomers, lower monomer content, and lower dosage. Product improvement should be sought through investigation of copolymers, bifunctional monomers, and various additives.

The commercial development program and the applied research program should be closely coordinated, so that both programs may be constantly redirected to provide the most pertinent data for specific end-use applications that appear to have the greatest potential. In determining the greatest potential, consideration should be given not only to the physical properties required by a given application but also to costs, since both are important in determining the feasibility of the application.

Continuing Work with WPC

At an information meeting on "Irradiated Wood Plastic Materials" sponsored by the Atomic Industrial Forum in cooperation with AEC on Sept. 15, 1965, in Chicago,* varied opinions were expressed by the participants. Although some companies are commercially producing developmental quantities of WPC, many of the speakers felt that more work needed to be done on economic and market evaluation, product testing, process refinement, physical-property evaluation, and basic research.

A number of projects in academic and industrial laboratories, some under AEC contract, are further evaluating and developing both the technical and economic aspects of WPC. Noteworthy among these is the continuing work at West Virginia University. In addition to the present program, work will be included on termite and decay resistance, paintability, and effectiveness of coloring wood by use of dyes incorporated into the monomers. A consolidation of recent economic and technical information and a design of a conceptual plant to provide meaningful cost projections are being prepared by Vitro Engineering Co. for the Commission. A program to test mechanical and physical properties is under way by the Research Triangle Institute and the Wood Products Laboratory of North Carolina State of the University of North Carolina. This will include perhaps 5000 individual tests to relate type of monomer as well as monomer loading to these properties as reflected by such tests as static bending, hardness, toughness, abrasion, compression, tension, and vibration. Among the woods studied will be northern red oak and yellow pine, representing heartwoods, and loblolly pine, representing sapwoods. The monomers studied will be principally methyl methacrylate and styrene/acrylonitrile.

Three independent regional studies to achieve steps toward commercialization of the process are being made under AEC–DID sponsorship by Southern Interstate Nuclear Board (Southeast), Pacific Northwest Laboratory of Battelle Memorial Institute (Northwest), and the Western New York Nuclear Research Center (Northeast).

A program to have industrial wood-product firms evaluate WPC in final product form has also been started. Under the plan, interested companies are to obtain, without charge, a limited amount of custom processing of their wood for subsequent fabrication of product samples for evaluation and testing. In return the participating firms will be required to pro-

*Proceedings of this meeting will be available about Mar. 1, 1966, from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

vide the Commission with a complete description of their tests and results, including ranking and effectiveness of polymers and treatments. An estimated 6 to 8 thousand pounds of wood will be involved in the program. On the basis of competitive bids submitted to the AEC, the Commission has selected the Lockheed-Georgia Company of Marietta, Georgia, a division of Lockheed Aircraft Corp., as the contractor to process these samples. The contractor will work out such details as sizes, shapes, and numbers of pieces with the selected firms and will advise them on the technical aspects of the process so that appropriate wood and plastic combinations can be selected which best meet the requirements of an individual product. The plan was announced July 16, 1965, and inquiries were requested by Aug. 20, 1965. The final selection of participating companies, made by the AEC on a representative basis, includes 78 organizations.

According to Kroehler Mfg. Co., there is still some question as to the use of polymer impregnation as a general method of treatment for all woods; i.e., the economics of solid woods may be too high. On the other hand, the possibilities in veneers could be very good. At present the surface of WPC veneer is not uniformly good, and cold pressing with standard urea resins is satisfactory but rather expensive. Impregnated solid woods may be quite suitable for certain portions of furniture, such as in legs and arms, to prevent scratching and nicking, and in glass doors for dimensional stability.

The Hardwood Plywood Manufacturers Association believes the use of WPC for ordinary plywood for paneling interiors is questionable at present. There may be a profitable future, however, in exterior hardwood plywood for outside finishes for siding, doors, stadium seating, and park benches.

Where cost of the starting material is only a small part of that of the finished products, WPC have perhaps their greatest potential. Hillerich & Bradsby Co. suggests that baseball bats, billiard cues, skis, golf clubs, etc., may very well have properties imparted to them by this process which will improve their qualities sufficiently to allow ready absorption of the added cost.

According to Rohm & Haas Co., methyl methacrylate seems to be the best polymer to date. They see no reason why polymer combinations could not involve hydrophobic monomers copolymerized with, for example, methyl methacrylate to impart moisture repellency.

Reductions in monomer price and source cost would favorably influence future product costs of WPC. The price of vinyl chloride recently was reported to have dropped from 8 cents per pound to 6.2 cents per pound.[69] To be effective for all WPC, other monomer prices must likewise drop.

The cost of ^{137}Cs was cut sharply[70] on Nov. 10, 1965. The new price for ^{137}Cs will be 12.5 cents per curie in lots greater than 200,000 curies—the current price for ^{60}Co is 50 cents per curie in large lots. Although more ^{137}Cs would be required for an equivalent source because of its lower gamma energy, single gamma emission per disintegration (2 γ's for ^{60}Co), and greater self-absorption, in the future source investment cost for ^{137}Cs may become competitive with ^{60}Co. The source depreciation cost for ^{137}Cs would be much less because of its much longer half-life, 30 years compared with 5.2 years for ^{60}Co. Of course, any reductions in ^{60}Co price* would also have a favorable influence on WPC cost where ^{60}Co is the source of radiation.

There are still many general questions as well as specific ones that need to be answered through rather extensive investigations. Work needs to be done on the uniformity of impregnation as well as the possibility of "case hardening" by impregnating a relatively thin layer of the wood surface prior to irradiation. It is still not completely understood exactly where the monomer goes and to what extent straight polymerization is involved as compared to graft polymerization. There are unanswered questions with respect to electrical properties of WPC, and more work needs to be done on the control of uniformity and quality of impregnations. However, it is apparent from this meeting that the proposed programs will answer many of these questions and that within a relatively short time WPC will fill its niche in the wood industry on the basis of the improvements imparted to wood commensurate with the incremental costs involved.

(R. E. Greene and P. S. Baker)

*See footnote on page 137.

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Section

IV

Isotopes and Radiation Technology

Source Development

Isotopic Power Data Sheets*

By S. J. Rimshaw†

Nuclear and physical properties of some unclassified radioactive source materials that have potential applications to terrestrial, marine, and aerospace power requirements were compiled from various publications.‡ The radioisotopes considered were ^{60}Co, ^{90}Sr, ^{137}Cs, ^{144}Ce, ^{147}Pm, ^{242}Cm, and ^{244}Cm. Experimental information on the properties of radioactive materials was very incomplete, and, where information was not available, the probable values were derived from data in the literature on inactive or related materials.

Cerium-144 as CeO_2

Isotopic composition	13.3% ^{144}Ce, 42.7% ^{142}Ce (stable), 44% ^{140}Ce (stable) after 1 year's decay
Half-life	^{144}Ce, 284.5 days; ^{144}Pr, 17.5 min
Decay and radiation properties	$^{144}Ce \rightarrow {}^{144}Pr \rightarrow {}^{144}Nd$ (stable) ^{144}Ce: β, 0.184 Mev (30%), 0.245 Mev (5%), 0.320 Mev (65%); γ, <0.134 Mev (30%) ^{144}Pr: β, 0.9 Mev (2%), 2.450 Mev (3%), 3.150 Mev (95%); γ, 2.18 Mev (2%), 0.695 Mev (3%)
Activity concentration	~1000 curies per gram of Ce metal at reactor discharge; ~440 curies per gram of Ce metal after 1 year's decay
Purity	Radiochemical, >99% ^{144}Ce (^{141}Ce activity is <1% of the ^{144}Ce after 1 year's decay); chemical, >99% CeO_2
Specific power	For 440-curie/g Ce metal, 2.84 watts per gram of CeO_2; thermal output of ^{144}Ce–^{144}Pr is ~7.9 watts/kc
Thermal energy	126 curies/thermal watt
Density	Theoretical, 7.3 g/cm³; measured, 6.6 g/cm³ (90%)
Power density	18.7 watts/cm³ from 440-curie/g Ce metal (6.6 g/cm³)
Thermal conductivity	For ThO_2, which is similar to CeO_2, varies from 0.0245 at 100°C to 0.0073 cal/(sec)(cm)(°C) at 1000°C
Expansion coefficient	10.7×10^{-6} to 12.9×10^{-6}/°C from 0 to 1310°C
Melting point	2680°C in air
Mechanical strength	No quantitative data
Thermal and radiation stability	CeO_2 undergoes decomposition and phase changes when heated in He atmosphere, finally yielding Ce_2O_3, which melts at 2190°C; effect of growing in of Nd_2O_3 on CeO_2 lattice not determined

(Continued on next page.)

*Presented at USAEC-sponsored Industry Information Meeting on Isotopic Power Development and Applications, Washington, D. C., May 18–19, 1964.

†Oak Ridge National Laboratory, Oak Ridge, Tenn.

‡Shielding data are taken from:

H. H. Van Tuyl, F. P. Roberts, and E. J. Wheelwright, Shielding Requirements for Promethium Sources, USAEC Report HW-77375, Hanford Atomic Products Operation, April 1963.

J. P. Nichols and E. D. Arnold, Radiation Characteristics and Shielding Requirements of Isotopic Power Sources for Space Missions, USAEC Report ORNL-TM-591(Rev.), Oak Ridge National Laboratory, August 1963.

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Radiation attenuation

Dose rate at 100 cm, rads/hr	Shielding, cm of U		
	100 watts	1000 watts	10,000 watts
100	0.9	3.4	5.8
10	3.5	6.1	8.6
1	6.2	9.0	11.5
0.1	9.0	11.8	14.2

Gas evolution due to radioactive decay: CeO_2 converted to Nd_2O_3; fate of excess oxygen unknown
Leach rate: No data
Vapor pressure: No data
Thermal-shock resistance: No data
Burnup characteristics: Dispersibility poor
Capsule compatibility: Reacts with Mo, Ta, W, Ir, and other refractory metals above 1400°C

Strontium-90 as $SrTiO_3$

Isotopic composition: 55% ^{90}Sr, 43.9% ^{88}Sr (stable), 1.1% ^{86}Sr (stable)
Half-life: ^{90}Sr, 27.7 years; ^{90}Y, 64.2 hr
Decay and radiation properties: $^{90}Sr \rightarrow {}^{90}Y \rightarrow {}^{90}Zr$ (stable)
^{90}Sr: β, 0.54 Mev (100%)
^{90}Y: β, 2.26 Mev (~100%)
Activity concentration: 33 curies per gram of $SrTiO_3$
Purity: Radiochemical, >99.9% ^{90}Sr + ^{89}Sr (^{89}Sr depends on decay and activity, usually <5% of the ^{90}Sr, at time of fabrication of source pellets); chemical, >95% Sr, rest chiefly Ca and Ba
Specific power: 0.223 watt per gram of $SrTiO_3$ (6.772 watts per kilocurie of ^{90}Sr)
Thermal energy: 148 curies/thermal watt
Density: Theoretical, 5.0 g/cm³; measured, averages 3.7 g/cm³ (3.2 to 4.2)
Power density: 0.825 watt/cm³ from 3.7-g/cm³ ^{90}Sr (^{89}Sr contributes at rate of 3.4 watts per 1000 curies)
Thermal conductivity: For nonradioactive $SrTiO_3$ varies from 0.0132 to 0.0173 cal/(sec)(cm)(°C) at room temperature, depending on density
Expansion coefficient: 1.12×10^{-5}/°C
Melting point: ~1900°C
Mechanical strength: Fair
Thermal and radiation stability: Thermal stability good; also radiation stability, as shown in 2-year-old samples

Radiation attenuation

Dose rate at 100 cm, rads/hr	Shielding, cm of U		
	100 watts	1000 watts	10,000 watts
100	0.07	0.7	2.0
10	1.0	2.3	3.8
1	2.6	4.3	5.8
0.1	4.4	6.2	8.0

Gas evolution due to radioactive decay: None
Leach rate: Nonradioactive $SrTiO_3$ leached at rate of 1 μg/(cm²)(day) in seawater; tests on 18-month-old fission-product $SrTiO_3$ pellets indicate a rate of ~1 mg/(cm²)(day)
Vapor pressure: No data
Thermal-shock resistance: No data
Burnup characteristics: Dispersibility poor
Capsule compatibility: Excellent with usual capsule materials such as stainless steel and Hastelloy C

Cesium-137 as Borosilicate Glass

Isotopic composition: 36.5% ^{137}Cs, 43.4% ^{133}Cs (stable), 20.1% ^{135}Cs (stable)
Half-life: ^{137}Cs, 30 ± 0.3 years; ^{137m}Ba, 2.6 min
Decay and radiation properties: $^{137}Cs \rightarrow {}^{137m}Ba \rightarrow {}^{137}Ba$ (stable)
^{137}Cs: β, 0.51 Mev (92%); 1.17 Mev (8%)
^{137m}Ba: γ, 0.662 Mev (92%); also isomeric transition
Activity concentration: 16 curies per gram of glass
Purity: Radiochemical, >95%, rest ^{134}Cs; chemical, >98% Cs, <2% Rb, small amounts of Na, K, SiO_2 from reagents
Specific power: 0.0774 watt per gram of glass (4.84 watts/kc)
Thermal energy: 207 curies/thermal watt
Density: 3.1 g/cm³ (~100%)
Power density: 0.24 watt/cm³ from 16-curie/g glass

Thermal conductivity	Estimated as 1×10^{-3} to 2×10^{-3} cal/(sec)(cm)(°C) at room temperature and 2.5×10^{-3} to 3×10^{-3} cal/(sec)(cm)(°C) between 1000 and 2000°C by analogy to sodium borosilicate glasses
Expansion coefficient	Estimated as 1×10^{-5} to 2×10^{-5}/°C (factor of ~4 less than ordinary glass)
Softening point	~1275°C
Mechanical strength	Brittle but no definitive measurements
Thermal and radiation stability	Devitrifies when held at 1000°C for a few hours

Radiation attenuation

Dose rate at 100 cm, rads/hr	Shielding, cm of U		
	100 watts	1000 watts	10,000 watts
100	1.5	3.4	4.7
10	3.6	5.5	7.2
1	6.0	8.1	9.8
0.1	8.4	10.8	12.6

Gas evolution due to radioactive decay	None
Leach rate	~0.2 mg/(cm²)(day) in H_2O at 30°C
Vapor pressure	Cs volatility from glass at 1200°C is ~3.5 mg/(cm²)(hr) [^{137}Cs, ~12 mc/(cm²)(hr)]
Thermal-shock resistance	Poor
Burnup characteristics	Does not oxidize; has ablative characteristics of a glass
Capsule compatibility	Excellent with usual capsule materials (stainless steel and Hastelloy C)

Promethium-147 as Pm_2O_3

Isotopic composition	~100% ^{147}Pm
Half-life	^{147}Pm, 2.67 years
Decay and radiation properties	$^{147}Pm \rightarrow {}^{147}Sm$ (stable) β, 0.225 Mev (100%)
Activity concentration	742 curies per gram of 6.6-g/cm³ Pm_2O_3
Purity	Radiochemical, ~1% ^{148}Pm ($T_{\frac{1}{2}}$ = 42 days) activity after 4 months' decay; after 2 years the ^{146}Pm ($T_{\frac{1}{2}}$ = 1.94 years) activity is 5×10^{-5}% of the ^{147}Pm, and the ^{148}Pm is even less; chemical, >95% ^{147}Pm; chief impurity at separation is Nd
Specific power	0.41 watt per kilocurie of ^{147}Pm (0.324 watt per gram of Pm_2O_3)
Thermal energy	2440 curies/thermal watt
Density	Theoretical, 7.3 g/cm³; practical, ~6.6 g/cm³ (90%)
Power density	2.03 watts/cm³ from 95% Pm_2O_3 (6.6 g/cm³)
Thermal conductivity	For typical rare-earth oxides, ~0.006 cal/(sec)(cm)(°C) at 150°C
Specific heat	For Sm_2O_3, 0.0944 cal/g from 0 to 1000°C
Expansion coefficient	For Sm_2O_3, 10.8×10^{-6}/°C from 30 to 740°C
Melting point	For Sm_2O_3, 2350°C
Mechanical strength	For Sm_2O_3 at room temperature: modulus of rupture, 2000 psi; modulus of elasticity (sonic), 26.5×10^6 psi
Thermal and radiation stability	Good for several years
Radiation attenuation	Little shielding required for ^{147}Pm; requirements established by ^{148}Pm and ^{148}Pm contents
Gas evolution due to radioactive decay	None
Leach rate	No data; most fired rare-earth oxides hydrate and disperse in boiling water
Vapor pressure	No data
Thermal-shock resistance	No data
Burnup characteristics	Dispersibility poor
Capsule compatibility	Excellent with most metals

Cobalt-60 Metal

Isotopic composition	31.6% ^{60}Co, 68.4% ^{59}Co (stable)
Half-life	^{60}Co, 5.24 years
Decay and radiation properties	$^{60}Co \rightarrow {}^{60}Ni$ (stable) β, 0.306 Mev (100%); γ, 1.17 Mev (100%), 1.33 Mev (100%)
Activity concentration	Maximum calculated as 360 curies/g for 7 years' irradiation at 2×10^{14} n/(cm²)(sec)*; 150 curies/g for 2 years' irradiation at 10^{14} n/(cm²)(sec)
Purity	Radiochemical, ~100% ^{60}Co; chemical, >99% except ^{60}Ni
Specific power	β, 0.22 watt/g; γ, 5.32 watts/g, based on specific activity of 360 curies/g (15.36 watts/kc)
Thermal energy	65.1 curies/thermal watt

*Editors' Note: Higher fluxes are now available, and the AEC is currently making some 700-curie/g material.

Density	8.7 g/cm^3 (cast metal)
Power density	48 watts/cm^3 (360 curies/g and $\beta + \gamma$ heating); fraction of the 46.2 γ watts absorbed depends on source size and geometry
Thermal conductivity	From 0.13 to 0.17 cal/(sec)(cm)(°C) at 30°C
Expansion coefficient	From 1.2×10^{-5} to 1.8×10^{-5}/°C at room temperature
Melting point	1480°C
Mechanical strength	Tensile, 34,000 psi; compression, 122,000 psi; Young's modulus, 30×10^6 psi
Thermal and radiation stability	Excellent in inert atmosphere

Radiation attenuation

Dose rate at 100 cm, rads/hr	Shielding, cm of U		
	100 watts	1000 watts	10,000 watts
100	3.7	5.5	7.0
10	5.7	7.5	9.1
1	7.6	9.4	11.0
0.1	9.5	11.3	12.9

Gas evolution due to radioactive decay	None
Corrosion rate	Nonradioactive: Dry air, ~0.00001 in./year; freshwater, ~0.0001 in./year; seawater, ~0.005 in./year Radioactive: Rapid in dry air, indicated from ORNL experience; penetration rate of at least 0.01 in./year and much higher in water
Vapor pressure	Boils at 2880°C at atmospheric pressure
Thermal-shock resistance	Excellent
Burnup characteristics	Oxidation rapid above 750°C
Capsule compatibility	Excellent
Heat of fusion	67 cal/g
Specific heat	~0.103 cal/(g)(°C)

Curium-242 as Cm_2O_3

Isotopic composition	>98% ^{242}Cm
Half-life	^{242}Cm, 162.5 days (spontaneous fission 7.6×10^6 years); ^{238}Pu, 89.6 years; ^{241}Am, 462 years
Decay and radiation properties	^{242}Cm → ^{238}Pu ^{242}Cm: α, 6.110 Mev (73.7%), 6.066 Mev (26.3%); γ, 0.441 Mev (26.3%)
Activity concentration	3044 curies per gram of $^{242}Cm_2O_3$; 1218 curies per gram of product (0.4 g of $^{242}Cm_2O_3$ + 0.6 g of $^{241}AmO_2$)
Purity	Radiochemical, >99%; chemical, 40% or less (60% ^{241}Am)
Specific power	44.1 watts per gram of Cm–Am oxides (~40% ^{242}Cm and ~60% ^{241}Am) (36.1 watts/kc)
Density	Theoretical, 11.2 g/cm^3; practical, ~9.0 g/cm^3 (~80%)
Power density	397 watts/cm^3 for 9-g/cm^3 $^{242}Cm_2O_3$ (40% Cm and 60% Am); 0.5% of total power contributed by ^{238}Pu and ^{241}Am
Thermal conductivity	0.0067 cal/(sec)(cm)(°C) at 125°C based on Gd_2O_3
Expansion coefficient	10.5×10^{-6}/°C at 25 to 1000°C based on Gd_2O_3
Melting point	1950°C in He
Mechanical properties	Modulus of elasticity (sonic), 14.5×10^6 psi at 25°C based on Gd_2O_3
Thermal and radiation stability	No data
Radiation attenuation	Gamma shielding required is small compared to neutron; neutron dose rate, 0.1 rad/hr at 50 cm for 2000-thermal-watt source with 10 cm of LiH shielding
Gas evolution due to radioactive decay	Source, originally 1000 curies, produces 13.9 cm^3 of He in 162.5 days
Leach rate	No data
Vapor pressure	No data
Thermal-shock resistance	No data
Burnup characteristics	Dispersibility poor
Capsule compatibility	No data

Curium-244 as Cm_2O_3

Isotopic composition	96.0% ^{244}Cm, 1.0% ^{245}Cm, 3.0% ^{246}Cm
Half-life	^{244}Cm, 18.1 years (spontaneous fission 1.4×10^7 years); ^{245}Cm, 8000 years; ^{246}Cm, 6600 years
Decay and radiation properties	^{244}Cm → ^{240}Pu (6.6×10^3 years) ^{244}Cm: α, 5.798 Mev (76.7%), 5.756 Mev (23.3%); γ, 0.0429 Mev (23.3%)
Activity concentration	72.6 curies per gram of $^{244}Cm_2O_3$
Purity	Radiochemical, >99%; chemical, ~99.5% with ~0.5% ^{243}Am
Specific power	2.53 watts per gram of Cm_2O_3 (34.28 watts/kc)
Thermal energy	29.2 curies/thermal watt
Density	Theoretical, 10.7 g/cm^3; practical, 9.0 g/cm^3

Power density	22.5 watts/cm^3 for 9.0-g/cm^3 Cm_2O_3
Thermal conductivity	0.0067 cal/(sec)(cm)(°C) at 125°C based on Gd_2O_3
Expansion coefficient	10.5×10^{-6}/°C at 25 to 1000°C based on Gd_2O_3
Melting point	1950°C in He
Mechanical properties	Modulus of elasticity (sonic), 14.5×10^{-6} psi at 25°C based on Gd_2O_3
Thermal and radiation stability	No data
Radiation attenuation	Neutron shielding required
Gas evolution due to radioactive decay	Source, originally 1000 curies, produces 84.7 cm^3 of He in 2 years (0.11 half-life)
Leach rate	No data
Vapor pressure	No data
Thermal-shock resistance	No data
Burnup characteristics	Dispersibility poor
Capsule compatibility	No data

Section

V

Isotopes and Radiation Technology

Isotopes Around the World

Economics of Worldwide Industrial Applications of Radioisotopes*

A survey conducted by the International Atomic Energy Agency (IAEA) indicated a total annual savings, in 26 countries, of $296 million to $400 million as a result of the industrial use of radioisotopes. Categories of uses were gaging, radiography, ionization, tracing (including activation analysis), massive irradiation, and miscellaneous. Industries were grouped into 12 classes.

Industrial applications of radioisotopes have developed primarily since the early 1940's. More than 1000 scientific papers were presented at 15 large national and international meetings, and at least 2500 original-research and survey papers were published on this subject in the decade ending with 1960.

The economic benefits derived from the use of radioisotopes are indicated by the increasing demand for them, but accurate dollar† values are few. The U. S. Atomic Energy Commission (AEC) in 1953 estimated total benefits of $122 million in the United States from such applications and in 1957, $400 million, using indirect numbers. A Soviet spokesman in 1958 gave a value of $170 million in the USSR. In 1959 the United States gave a new value of $157 million based on a more detailed survey, and subsequent USSR estimates have been lower. However, United Kingdom and Swedish surveys have indicated considerable direct, indirect, and potential benefits.

In 1963 the International Atomic Energy Agency (IAEA) published a literature survey[1] of radioisotope applications described in the scientific literature up to 1960, classified according to industry. However, information was insufficient to show the extent of the use of radioisotopes and the derived economic benefits, and the IAEA decided to survey the field. Questionnaires (Fig. V-1) on industrial uses of radioisotopes were sent to member countries, 25 of whom eventually participated in the survey. The answers were considered confidential. The USSR did not participate, but a few of their known figures were included. Each country filled in the questionnaire according to its own interpretation.

The data obtained from the survey were studied by experts from various countries at a meeting in Vienna on March 16 to 20, 1964. For purposes of analysis, industries were split into 12 broad groups: food; tobacco; textiles and footwear; wood and paper; leather and fur; rubber; chemicals and plastics; cement, glass, and china; petroleum and coal; basic metals; machinery; and services. Applications were considered under the headings: gaging, radiography, ionization, tracing, massive irradiation, and, of course, miscellaneous. Forty participants from 22 countries, 23 observers from seven nonparticipating member states and three international organizations, experts in statistics and economics, and well-known scientists who reviewed the various techniques concluded that industrial applications of radioisotopes have resulted in the following savings:

	Millions of dollars			
Application	USA (1963)	USSR (1961)	24 other countries (1961–1963)	Total
Gaging	$ 35–50	$100	$27–43	$162–194
Radiography	4.0–7.6	22	12–29	38–58
Ionization	Included in other groups		1–2	1–2
Tracing	27–48	58	10–40	95–146
	$ 66–106	$180	$50–114	$296–400

*Based on *Industrial Radioisotope Economics*, Findings of Study Group Meeting, Vienna, 1964, International Atomic Energy Agency, Vienna, 1965 (STI/DOC/10/40), 576 + iii pp., $11.50.

†All dollar values quoted in this report are U. S. dollars.

INTERNATIONAL SURVEY ON THE USE OF RADIOISOTOPES IN INDUSTRY

Company's name and address of the establishment where the isotopes are used:

Please complete a separate questionnaire for each broad product group in the processing of which isotopes were used in 1961.

Name of broad product group: _____

PART I: Manufacturing savings

1. (i) Total value of output in 1961	
(ii) Value of isotope assisted output 1961	
2. In what year did you first use radioisotopes in processing the product group mentioned?	
3. What was the cost (or if not available, the present book value) of all isotopes and related equipment and facilities at present in use?	
(i) cost of isotopes	
(ii) cost of isotope equipment (such as gauges, counters, etc.)	
(iii) cost of health physics equipment	
(iv) cost of structures especially required for radioisotope use, including installation costs	
(v) other	
4. What was the cost of all isotopes and related equipment and facilities you installed or acquired in 1961?	
5. What was the yearly cost of the maintenance of your isotope equipment in 1961, including health precautions (depreciations not included)?	

6. Show by ticks in appropriate boxes where cost savings have been achieved by radioisotope techniques. If possible, also give approximate amount of annual cost savings. *

- (i) raw material ☐
- (ii) product scrap ☐
- (iii) labour ☐
- (iv) other, including savings from lower capital costs or maintenance or increased productivity in general ☐

* *If costs have increased, show amount of increase, in brackets. If you can only give a total figure of savings, please indicate how this has been derived (e.g. as percentage of turnover). If you cannot give cost figures, kindly indicate the magnitude in terms such as tons of scrap, man-hours, etc. (Please use back page if necessary).*

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PART II: Check list of radioisotope techniques and benefits

Please indicate by ticks in the appropriate boxes the techniques you employ, and the ways in which they are superior to previous methods.

TECHNIQUES	BENEFITS (show cost savings in Part I, Item 6)
A. *Radioisotope gauging*	
Thickness ☐ Density ☐	Product of better quality ☐
Level ☐ Component analysis ☐	Closer control within tolerances ☐
Other, please specify:	Saving of raw material ☐
	Reduction of rejects ☐
	Saving of scrap ☐
The gauging is applied to:	Saving of labour ☐
Is the use regular ☐	Others, please specify:
occasional ☐	
exceptional ☐	
Sources: number, strength and nuclide:	
B. *Industrial radiography*	
Inspection of welds ☐, castings ☐,	Better control of raw materials ☐
materials to be machined ☐	Better control of products ☐
Other, please specify:	Reduced time for inspection ☐
	Others, please specify:
Is the use regular ☐	
occasional ☐	
exceptional ☐	
Sources: number, strength and nuclide:	
C. *Ionization applications*	
Static eliminators ☐	Increased speed of production ☐
Ionization sources in electron tubes, etc. ☐	Reduced product scrap ☐
Other, please specify:	Reduced fire hazard ☐
	Others, please specify:
Is the use regular ☐	
occasional ☐	
exceptional ☐	
Sources: number, strength and nuclide:	

2

Fig. V-1 English version of questionnaire sent by IAEA to member countries as part of its international survey on industrial uses of radioisotopes.

(This figure continues on the next page.)

TECHNIQUES	BENEFITS (show cost savings in Part I, item 6)
D. *Radioactive tracers in research process and product control*	
Wear and corrosion studies ☐	Clearer understanding of processes ☐
Process plant studies ☐	Better control of process operation ☐
Leak detection ☐	Better control of product ☐
Chemical reaction studies ☐	Lower cost of analysis ☐
Chemical analysis ☐	Improved plant efficiency ☐
Other, please specify: _____	Others, please specify: _____
_____	_____
Is the use regular ☐	_____
occasional ☐	_____
exceptional ☐	
Sources: nuclides _____	
E. *Massive irradiation*	
Sterilization of medical and pharmaceutical supplies ☐	Please state benefits:
Sterilization of foods ☐	
Promotion of chemical reactions ☐	
Other, please specify: _____	
_____	
Is the use regular ☐	
occasional ☐	
exceptional ☐	
Sources: number, strength and nuclide: _____	
F. *Miscellaneous applications*	
Luminescence ☐ Atomic batteries ☐	Please state benefits:
Other, please specify: _____	
_____	
Is the use regular ☐	
occasional ☐	
exceptional ☐	

3

PART III: Further detailed description of experience with radioisotopes

In order to make possible presentation of some detailed case examples of how radioisotopes have been of benefit, we would appreciate any more detailed description of your experience you would care to present below. For example we would be interested to know whether radioisotopes have enabled you to do something not possible before.

PART IV: General comments and additional information

_____ _____ _____

Name and title of official *Signature* *Date*

4

Fig. V-1 (Continued.)

The gaging values are considered reliable, the differences between low and high estimates reflecting the method used in scaling up results to national and international levels rather than a high uncertainty in the basic values. Values for radiography are less reliable since few countries contributed useful data. The economic importance of ionization appeared to be low, and economic information on tracer methods was very limited. No estimates were made for massive-irradiation and miscellaneous categories since these applications had so far not resulted in comparable savings.

The final report on the study-group meeting presents the data first by country and then by technique. Only the technique portion was used in the review presented below.

Gaging

Gaging accounts for half the total number of all industrial radioisotope applications. Because radioisotope gages make measurements without any change in, or contact with, the material on which measurements are made, they may be used with continuously produced materials at high temperatures or traveling at high speeds, such as hot steel or plastic; malleable or soft sheet, such as paper; and materials where surface finish is important, such as precious metals. Because of the penetrating ability of gamma rays, externally mounted sources and detectors can be used to measure densities and levels of solids, liquids, and slurries inside pipes, tanks, and sealed containers where abrasive or corrosive conditions may, or sanitary conditions must, prevail. Radioisotope sources are compact, they require no electric-power supply, and their energies and intensities are stable. The absence of moving parts, the external mounting, and their ease of feedback contribute to the economies derived from their use. Most radioisotopes used in gages (Table V-1) have long lives and need be replaced only infrequently.

A large number of items in everyday use have been produced with the aid of radioisotope instruments. Processing and packaging of many foods are controlled with radioisotope gages; the high quality of modern tires is partly due to the use of gages; and density and level gages are widely used in cement, glass, and petroleum production. Most sheet-metal rolling mills are equipped with radioisotope instruments, and production of tinplate and galvanized iron is controlled with X-ray-fluorescence and beta-backscatter gages. Construction of dams, buildings, roads, and airfields is monitored with the aid of radioisotope density and moisture gages. Machine builders accept radioisotope gages as standard instruments and now frequently provide for them on new machinery.

Table V-1 RADIOISOTOPE SOURCES USED IN GAGES

Country	Radioisotope	No. of sources	Total activity, mc
Federal Republic of Germany	^{60}Co	622	13,182
	^{137}Cs	43	6,071
	^{198}Au	1	7
	^{170}Tm	1	200
	^{204}Tl	248	1,157
	^{90}Sr–^{90}Y	102	1,518
	^{147}Pm	19	1,425
	^{85}Kr	18	459
	^{107}Ru–^{107}Rh	2	50
	^{226}Ra	9	66
Netherlands	^{60}Co	111	
	^{137}Cs	46	
	^{204}Tl	18	
	^{90}Sr–^{90}Y	88	
	^{147}Pm	-	
	^{85}Kr	30	
South Africa	^{60}Co	8	656
	^{137}Cs	5	560
	^{192}Ir	4	400
	^{204}Tl	19	350
	^{90}Sr–^{90}Y	61	1,120
	^{226}Ra–Be	4	32
United Kingdom	^{60}Co	84	
	^{137}Cs	40	
	^{134}Cs	1	
	^{192}Ir	1	
	^{24}Na	1	
	^{170}Tm	10	
	^{204}Tl	155	
	^{90}Sr–^{90}Y	115	
	^{147}Pm	8	
	^{85}Kr	14	
	^{144}Ce–^{144}Pr	8	
	^{106}Ru–^{106}Rh	2	
	^{14}C	1	
	^{3}H	2	
	^{226}Ra	6	
	^{210}Pb	2	
	^{210}Po	1	

Gages are classified here according to the property measured: thickness and mass per unit area, density, level, and components (Table V-2). The number of gages used in industries of various countries is shown in Table V-3.

Variations from one country to another in the relative number of instruments in a given industry are due principally to the different major industries in each country. For example, in Finland 72% of the radioisotope gages are in the paper industry as compared with 18% in the United Kingdom. On the other hand, there are 897 tobacco gages in the United Kingdom, 59 in the Netherlands, and none in France and the

Table V-2 APPLICATIONS OF GAGES IN VARIOUS INDUSTRIES

Industry	Type of gage	Application
Food, beverage	Thickness; mass per unit area	Sheets of dough and thin slabs of chocolate
	Density	Air content of ice cream; fruit juice; syrup; condensed milk; tomato paste; fat content of baby food; ground corn slurry (starch and gluten production)
	Level	Soup, meat, beans, coffee, beer in cans, packets, and bottles; limestone, coke in lime kilns (sugar refining); sugar and chemicals in storage vessels, hoppers, reaction vessels, and fermenting tubs; grain in bins; evaporated grain syrup (whiskey making)
Tobacco	Density	Cigarettes
	Level	Finished product in packages
Textile, clothing, footwear	Thickness; mass per unit area	Thread; warp-knit fabrics (heat setting); coatings on cellulosic fabrics, tufted carpets, and leather cloth; tire cords; adhesive-coated surgical dressings; linoleum; footwear materials; felt and fabric wear
	Density	Polymer, synthetic yarn solutions; sulfuric acid
	Level	Reaction-vessel contents
Wood, paper	Thickness; mass per unit area	Paper, paperboard, chipboard, and pulp; coated, laminated paper; coatings, ink on paper
	Density	Timber; straw building board; digester liquors; clay slurries and solutions; black-liquor evaporator feeds
	Level	Wood chips, pulp in containers, reaction vessels, and digesters; chlorine levels in bleaching towers
Rubber	Thickness; mass per unit area	Rubber sheet on calenders; rubber goods; foam rubber; rubber coating of tire cords; coated fabrics; tires; floor covering
	Density	Latex solutions; butadiene and styrene (degree of polymerization)
	Level	Powder in packets
Chemicals, plastics	Thickness; mass per unit area	Plastic sheets; tapes; coated metal; paper and fabrics; laminated products; adhesives; walls of plastic bottles, pipes, and tanks
	Density	Process solutions, brine, salts, acids, detergents, plastic and organic solutions, slurries, and explosives
	Level	Liquids, solids, liquefied gases, slurries in containers, reaction vessels, sulfuric acid, CO_2, SO_2, asphalt, coal, coke, lime, cement, explosives, plastics, and catalysts; contents of bottles, tubes, and packets
	Analysis	Chlorine, sulfur content of plastics
Glass, mineral, cement	Thickness; mass per unit area	Abrasive-coated paper, textiles; glass, asbestos fibers; asbestos, cement panels
	Density	Cement, lime, asbestos, and mineral-ore slurries; mineral-wool insulation; moisture
	Level	Molten glass in furnaces; lime, sand, and cement slurries; clinkers; plaster
	Analysis	Mineral ores (X-ray fluorescence); boron, lead in glass; fire-bricks (scintillation scanning)
Petroleum, coal	Thickness, mass per unit area	Coal seams; asphalt-sheet products (roofing paper, shingles); cement slurries in wells; walls of pipes, tanks
	Density	Petroleum, petroleum fractions; sand-brine; coal; coke; catalysts; sulfuric acid (alkylation); bulk density of coal in bunkers
	Level	Coal, coke in bunkers, hoppers, and skips; catalysts, liquids in reaction vessels; hydrocarbons in cracking units, trays in distillation columns; petroleum in delayed coking units; hot oil in melting tanks; alignment of machines in coal, coke preparation; propane, butane levels in cylinders
	Analysis	S, Co, Pb, H_2S contents; C/H ratios; ash
	Various	Logging
Basic metals	Thickness, mass per unit area	Hot- and cold-rolled sheet metal; tubes; coated-, laminated-metal products; tinplate, galvanized iron; walls of pipes, tanks; moisture in powdered, slurried ores, other charge materials, and foundry sand
	Density	Powdered, slurried ores
	Level	Coal, metal, ores in storage bunkers, hoppers; liquid metals in cupola, blast furnaces and crucibles, molds; loads in electro-thermal kilns, furnaces; dust in electrostatic precipitators; slurries; sodium in tubes; ores, leaching solutions in pressure vessels
	Analysis	Metal (Cu, Fe, etc.) ores (X-ray fluorescence)
Machinery	Thickness; mass per unit area	Paper, rubber, plastic, textile, metal sheets, tapes, coated and uncoated; condenser paper; carbon steel; rubber compound for batteries; walls of pipes, tubes
	Density	Latex, condensed milk, and pulverized coal
	Level	See basic metals
Services	Density	Silt in dredging; gas, steam, steam–water, fluidized coal, sand, concrete, and cement; soil, moisture
	Level	Liquids, liquid gases in containers; water in steam boilers

Table V-3 NUMBER OF GAGES USED IN INDUSTRIES OF VARIOUS COUNTRIES*

Country	Year	No. of gages												
		Food, drink	Tobacco	Textiles, clothing, footwear	Wood, paper	Rubber	Chemicals, plastics	Glass, minerals, cement	Petroleum, coal	Basic metals	Machinery	Services	Not identified	Total
Argentina	1963			2T	3T		1T	1L	20 log	4T 1L		1T		33
Australia	1962	1D	32D	3T	12T		20T 1D 9L	1D	30L 3V	2T 4L	2L 1V	2L 1V		124
Austria	1961–1962				16T	5T	7T 1L		3V	2T				34
Belgium	1962–1963	4L	4D	2T	42T 5L	1T	9T 10L	6L	1T 1D 26L 1V	9T 20L		3L		144
Canada	1963	1T 13D 10L	464D	1T	165T 16D 12L	8T	17T 16D 85L	1T 11D 9L	9T 4D 6L 203 log	17T 135D 12L 36V	43T 9D 30L	22T 35D 4L		1394
Czechoslovakia		1D 1L		2T	5T	3T	8T 6D 15L	5L	2D	58T 2L	2L	5T		115
Denmark	1963	3T	72D	6D	56T	2T	5T 6L	2D	1D 6L 1CA	1T 6L	2L	5L		174
Finland	1961–1962			2T	31T 17L	1T	3T 1D 5L		4L	2D 2L		1T		69
France	1961	7L		81T 6L	166T 11L 1V	35T	38T 15D 102L 8CA	2T 40L 1CA	8D 125L 10V 94 log	16T 2D 164L	20T 7D 119L 15V	279L 27V	16T 16D 5L 5CA 24V	1465
Federal Republic of Germany	1962–1963	1T 1D 2L		18T	382T 5D 5L	69T 1D	427T 5D 22L	86T 16L	63T 2D 7L	198T 1D 20L	20T 1D 20L	11T 1D 7L	3T	1375
Japan	1961			6T 42L	20T 26L 3V	11T	13T 2D 56L 6V	8T 1D 1L	1T 2D 7L	46T 7D 9L 3V	14L 4D 10L 2V	5T 9D 2L 11V	3T 6D 1L 4V	341
Netherlands	1962		59D		63T	10T	11T 3L	9L	128L	5T 1L		7D 8L		304
Norway			3D	5T	48T 9L	4T	6T			2T 1D 3L	1D 2L			84
Poland	1962				5T	4T	1D 44L	10L 1 log	3L 33L(sic) 8 log	8T 3L 1CA		98L		219
Portugal	1962				3T		1V		1L 1V			2V		8
South Africa	1961		59D		10T	11T	1D 1V	2L		12D 2L 2V		3V	10V	113
Spain	1962		2D		4T	6T	12T 2L			12V		2V	1V	41
Sweden	1963	10L	20D	20T	140T 1D 110L	10T	75T 30L	10T	4D 5L	10L	15T 20L			480
United Kingdom	1961	1T 2D 16L	897D	72T	357T 3D 2L 1CA	41T	71T 25D 161L 1CA	17T 2D 34L 1CA	4T 8D 20L 19CA	93T 1D 45L 3V	51T 11D 27L 2CA 4V	11T 9D 12L 2CA 12V		2038
USA	1957 1964													4000 8000– 9000
Yugoslavia		1D 4L		1T	3T 2D 3L	2D	1V 1D 7L	1T	2V	8T 3V	1T 1V			41

*T, thickness or mass per unit area; D, density; L, level; V, various; CA, component analysis; log, logging.

Federal Republic of Germany, but the number of cigarettes produced in the last three countries in 1961 was 13, 47, and 56×10^9, respectively. The apparent discrepancy here may be due to raw tobacco being relatively cheap, and savings therefore small. Nevertheless, the success achieved with beta gages in producing uniform-weight cigarettes should eventually lead to their adoption by all manufacturers. Other variations may be explained by good salesmanship or ready availability of particular instruments.

Variations from one industry to another in the type of gage that is predominant reflect chiefly

the number of processes to which radioisotope gages can be applied. There are more level and density gages in the petroleum and coal and in the basic-metal industries and more thickness gages in the wood and paper industries.

ECONOMICS

The annual savings from applications of radioisotope gaging in world industry range from $162 million to $194 million yearly (Table V-4). These values reflect only direct savings to industry itself and do not include customer or intangible savings or savings to the national economy from better allocation of resources.

Table V-4 ESTIMATED WORLD SAVINGS RESULTING FROM USE OF RADIOISOTOPE GAGES

Country	No. of gages	Estimated annual savings, dollars	
		Low	High
Argentina	33	$ 200,000	$ 400,000
Australia	125	500,000	600,000
Austria	34	120,000	180,000
Belgium	144	520,000	840,000
Canada	1,394	800,000	2,400,000
Czechoslovakia	115	1,000,000	1,500,000
Denmark	174	300,000	1,000,000
Finland	69	750,000	900,000
France	1,465	2,600,000	3,800,000
Federal Republic of Germany	~2,000	5,000,000	8,000,000
Japan	341	2,000,000	3,000,000
Netherlands	304	1,000,000	1,500,000
Norway	84	300,000	300,000
Poland	219	1,900,000	2,900,000
Portugal	8	-	-
South Africa	113	200,000	300,000
Spain	41	150,000	300,000
Sweden	480	1,800,000	3,000,000
United Kingdom	2,037	7,500,000	12,000,000
Yugoslavia	41	125,000	400,000
		~26.7 million	~43.4 million
USA	~9,000	35.2 million	50.4 million
USSR	?	100 million	100 million
		~162 million	~194 million

The economic benefits derived from the use of radioisotope gages result from a number of factors. Capital, operation, and maintenance costs are lower than those of other methods offering the same technical benefits. Sophisticated health physics instrumentation is rarely needed, and insurance premiums are comparable with those for other plant instrumentation of equivalent value. The use of radioisotope gages—the only kind that can be used for some materials—may decrease the man-hours necessary for monitoring a process, accelerate analytical work, and increase plant efficiency where conventional process gaging is a bottleneck.

Savings in raw material and scrap are important, particularly when the raw-material cost is a significant fraction of the final-product cost. An accurate gage often allows the amount of material used to be kept within narrow limits and thus decreases both the raw material per unit product and the amount of scrap. Increased quality, the most quoted advantage of nuclear gaging, makes the product more competitive although no direct benefits are obtained.

Radioisotope instruments range in price from about $1500 for a rudimentary basic model (source, detector, and indicating unit) to $20,000 or more for a refined instrument engineered to operate continuously and accurately and to indicate material quality under prevailing conditions. Some provide automatic standardization, computer interpretation of detector output, and automatic process control. Gages in the low-price range include simple on–off switches for indicating, for example, when powdered soap in a carton reaches a required level. They consist of a low-activity source, Geiger counter, and rate meter unit operating a relay. When a wider range of levels must be covered or measurements made on larger containers, the price is higher because of increase in source activity, number of detectors, and other factors.

For continuous, accurate, reliable measurement of, for example, mass per unit area on a continuously produced material, rudimentary components alone are practically useless. They must be rigidly mounted to maintain the desired geometry. Since the mounting will be subject to jarring and may be used by operators to climb on the machine, sturdy construction is required. Allowances must be made for accumulation of dust, temperature variations, source decay, and material-composition changes. For scanning across a web, elaborate construction is required.

Radiography

Gamma radiography is a widespread and generally accepted method of nondestructive testing; it depends on the difference in absorption of gamma rays by materials of different densities or thicknesses. In some cases gamma sources, which require no electrical connections, provide the only means of radiography—for shapes not amenable to X-ray tube use, for inaccessible

locations such as ship's welds in a hull, in industrial plants where explosions could result from release of electrical charges by high-voltage installations, and in remote areas. It may be the only applicable testing method for flaws, cracks, or inhomogeneities in castings or ingots or for custom-built objects. As the radiation from a radioisotope source is emitted in all directions, large areas can be covered in a single exposure, while X-ray tubes would require many exposures, a longer total exposure time, and more working hours. Cobalt-60 sources are comparable with radium and highly penetrating machine X-ray sources for very thick materials, e.g., 40-mm steel, although with thinner materials the images produced are of lower quality than those produced by X rays. Gamma radiography, depending on the source, may require longer exposures than X-ray radiography.

Neutron radiography, with generators or with isotope sources, is used in a few specialized applications; for example, it can detect a paraffin inclusion in 30 cm of lead. Bremsstrahlung radiography has not been extensively used because of the source dimensions required. The best physical practice competitive with radiography, ultrasonics, does not give so satisfactory a record and is not applicable to so many problems.

Nuclides in common use for gamma radiography are ^{60}Co, ^{137}Cs, and ^{192}Ir; others sometimes used for special purposes are included in Table V-5.

Table V-5 ISOTOPES SUITABLE FOR GAMMA SOURCES

Isotope	Half-life	Gamma energies, Mev
^{60}Co	5.3 years	1.2, 1.3
^{154}Eu	5 years	0.12, 0.34, 0.41, 1.2
^{182}Ta	120 days	0.15, 0.22, 1.13, 1.22
^{124}Sb	60 days	0.12, 0.61, 0.65, 0.73
^{46}Sc	85 days	0.88, 1.12
^{192}Ir	70 days	0.3, 0.47, 0.60
^{75}Se	127 days	0.12, 0.14, 0.27, 0.4
^{181}Hf	46 days	0.48, 0.34
^{141}Ce	28 days	0.14
$^{144}Ce-^{144}Pr$	275 days	0.22, 1.2
$^{137}Cs-^{137m}Ba$	30 years	0.662

The number of gamma radiography sources used in industries of various countries is shown in Table V-6. Food, tobacco, and leather industries did not report any.

ECONOMICS

The estimated total world savings resulting from the industrial use of gamma radiography are $58 million (Table V-7). A large part of the direct savings by firms that replaced X-ray machines and radium sources with artificial radioisotopes accrued from lower equipment and maintenance costs or elimination of radium rental fees. Other factors were decrease in labor costs, increase in inspection productivity, decrease in raw materials and scrap, and in-

Table V-6 NUMBER OF GAMMA RADIOGRAPHY SOURCES IN INDUSTRIES

Country	Textile	Wood, paper	Rubber	Chemicals, plastics	Cement	Petroleum, coal	Basic metals	Machinery	Services	Not identified	Total
Argentina	–	–	–	1	–	5	2	1	18	–	27
Australia	–	–	–	–	–	1	12	7	30	–	50
Austria	–	–	–	–	–	–	30	–	–	–	30
Belgium	–	–	–	–	–	1	17	13	20	–	51
Canada	–	–	–	–	–	5	60	45	35	–	145
Czechoslovakia	–	–	–	–	8	–	12	138	10	–	168
Denmark	–	–	–	–	–	–	1	–	20	–	21
Finland	–	–	–	–	–	–	4	1	3	–	8
France	–	–	1	11	1	19	80	300	50	38	500
Federal Republic of Germany	–	1	–	10	15	6	40	200	28	–	300
Japan	1	–	–	8	–	–	39	112	2	8	170
Netherlands	–	–	–	–	–	8	–	127	–	–	135
Norway	–	–	–	–	–	1	1	13	1	–	16
Poland	–	–	–	–	–	–	–	60	20	–	80
Portugal	–	–	–	–	–	–	–	10	–	–	10
South Africa	–	–	–	1	–	–	2	14	31	–	48
Spain	–	–	–	–	–	–	1	16	–	–	17
Sweden	–	–	–	–	–	–	10	20	50	–	80
United Kingdom	–	–	–	14	4	10	147	364	167	–	706
Yugoslavia	–	–	–	–	–	1	3	93	25	2	124
	1	1	1	45	28	57	461	1534	510	48	2686

crease in net profits. Indirect benefits include better product quality and improved customer relations.

Some specific examples of savings taken from the national reports are: Replacement of X-ray machines and radium sources with artificial radioisotopes in 15 companies decreased annual equipment and facility costs from $200,000 to $29,000. Expenditure of $8800 for a radioisotope unit, compared to $119,000 for an X-ray unit giving the same benefits, was accompanied by a decrease of $24,000 in labor costs because of increased portability and capability of the isotope source. The purchase of several hundred dollars' worth of ^{60}Co eliminated an annual rental fee of $30,000 for radium.

Table V-7 ANNUAL WORLD SAVINGS ESTIMATED FOR GAMMA RADIOGRAPHY

Country	No. of sources	Savings reported	Estimated annual savings Low	Estimated annual savings High
Argentina	27	-		
Australia	50	$ 66,000		
Austria	30	-		
Belgium	51	-		
Canada	145	-		
Czechoslovakia	168	-		
Denmark	21	-		
Finland	8	-		
France	~500	4,000,000	$ 2,000,000	$ 8,000,000
Federal Republic of Germany	~300	12,000		
Japan	170	4800–6500	850,000	1,500,000
Netherlands	135	700,000	700,000	700,000
Norway	16	-		
Poland	80	800,000	400,000	800,000
Portugal	10	-		
South Africa	48	-		
Spain	17	33,000*		
Sweden	80	100,000	100,000	200,000
United Kingdom	706	3,700,000*	5,700,000	11,400,000
Yugoslavia	124	$ 200,000*	500,000	1,500,000
		Subtotal	~10.3 million	~24.1 million
16 other countries	(900)		1.8 million	4.8 million
USA			4.0 million	7.6 million
USSR			22 million	22 million
		Grand total	$38 million	$58 million

*Values are incomplete.

Ionization

Radioisotope applications based on interaction of radiation with gases are useful in various industrial and scientific problems, but world industry has not adopted them uniformly (Table V-8). The annual saving resulting from their use is conservatively estimated as $1 million to $2 million, exclusive of USSR figures.

The use of instruments with radioactive isotopes for smoke detection is now approved by fire insurance companies in several countries. Radium-226 was used in early designs but has been replaced by ^{241}Am. Gas chromatographic analysis with instruments using radioisotope sources ($^{90}Sr-^{90}Y$, ^{147}Pm) has become a very sensitive method since the introduction of high-sensitivity detectors. With the argon detector (Table V-9), multicomponent mixtures of only a few micrograms can be separated. Examples of uses are analysis of mine gas and pesticides and determination of acetylene in oxygen-production plants. Radioisotopes (Table V-10) can significantly decrease the ignition delay time in cold-cathode discharge tubes. And mass flow or linear velocity of gases moving at high speed may be measured by radioisotope-induced ionization without impedance to the flow. Less well-developed applications are lightning conduction, static-charge elimination, and vacuum gaging.

Tracing

Annual world savings resulting from industrial tracer uses (Table V-11) are roughly es-

Table V-8 NUMBER OF IONIZATION SOURCES IN INDUSTRIES*

Country	No. of sources												
	Food	Tobacco	Textile	Wood, paper	Rubber	Chemicals, plastics	Cement	Petroleum, coal	Basic metals	Machinery	Services	Not identified	Total
Belgium				2		1							3
Canada												1022	1022
Czechoslovakia			2		2	6		6					16
Denmark			41			4							45
Finland			7	1									8
France			3	2		4				8		7	24
Japan									12	22			34
Netherlands				4		12							16
Norway				1	2	1							4
Poland					18								18
South Africa	1											10	11
Spain						9					49		58
United Kingdom	15	6	58	12		223	4	69	1	46	38		472
Yugoslavia		2	3		2	2				1		1	11
	16	8	114	22	24	262	4	75	13	77	87	1040	1742

[*Text indicates uses in USA, but these were not included in the table in the original report.]

Table V-9 PERFORMANCE OF DETECTORS USED IN GAS CHROMATOGRAPHY

Detector	Carrier gas	Background current, amp	Minimum impurity detectable, g	Detectable substances
Cross-section	H_2	10^{-9}–10^{-8}	2×10^{-7}, propane	All
Argon	Argon	10^{-8}	4×10^{-13}, propane	Most organic and inorganic vapors
Electron-capture	Argon	3×10^{-9}	3×10^{-14}, CCl_4	Halogen and oxygen compounds
Electron-mobility	Argon	3×10^{-10}	10^{-11}, CO_2	Permanent gases and organic vapors

Table V-10 PROPERTIES OF RADIOISOTOPES USED TO CONTROL IGNITION DELAY TIMES IN COLD-CATHODE DISCHARGE TUBES

Isotope	Half-life	Energies of emitted radiations, Mev		
		α	β (max.)	γ
^{3}H	12.3 years	-	0.018	-
^{85}Kr	10.6 years	-	0.67 (99.6%), 0.15 (0.4%)	0.51 (0.4%); also bremsstrahlung
^{63}Ni	125 years	-	0.067	-
^{226}Ra and decay products	1760 years	Many, 4 to 8	Many, 0.01 to 3.0	Most abundant: 0.6, 1.12, 1.76
$^{90}Sr-^{90}Y$	28 years, 2.7 days	-	0.54 to 2.25	-
^{232}U and decay products	74 years	Many, 4.5 to 8.8	Chiefly 0.05, 0.34, 0.58, 1.55, 2.25	Miscellaneous, below 0.3

Table V-11 NUMBER OF TRACER USERS IN VARIOUS INDUSTRIES

Country	No. of users												
	Food	Tobacco	Textile	Wood, paper	Rubber	Chemicals, plastics	Cement	Petroleum, coal	Basic metals	Machinery	Services	Not identified	Total
Argentina	-	-	-	-	-	-	-	2	-	-	4	-	6
Australia	-	-	-	1	-	2	1	1	1	1	3	2	12
Austria	-	-	-	-	-	2	-	1	-	-	2	-	5
Belgium	-	-	-	-	-	5	-	-	2	-	-	-	7
Czechoslovakia	-	-	3	1	3	6	4	4	8	6	3	-	38
Denmark	1	-	-	1	1	8	1	-	-	2	-	-	14
France	-	3	7	-	3	38	7	33	15	28	19	57	210
Federal Republic of Germany	-	-	-	-	-	14	3	7	24	16	1	-	65
Japan	-	-	6	2	-	12	3	3	14	15	3	-	58
Norway	-	-	1	-	-	2	-	-	-	1	1	-	5
Poland	-	-	-	-	-	2	2	1	9	-	1	-	15
Portugal	-	-	-	-	-	-	1	-	-	-	1	-	2
South Africa	-	-	-	-	-	1	-	-	8	-	-	-	9
Spain	-	-	-	-	-	1	-	-	-	-	1	-	2
Sweden	-	-	-	7	-	6	1	-	8	3	3	-	28
United Kingdom	5	-	1	2	1	41	3	9	14	25	25	-	126
USA (1958)	14	3	-	5	3	135	14	156	22	126	82	-	560
Yugoslavia	3	-	1	-	-	6	-	2	4	6	-	6	28
	23	6	19	19	11	281	40	219	129	229	149	65	1190

timated as $95 million to $146 million. Activation analysis is included in this category.

Radioisotopes are used as tracers in various phases of industrial work. In process control, optimum working conditions can be determined by following a tracer through a process. In product control, labeled units facilitate inspection, and fabricated vessels can be leak-tested, e.g., with ^{85}Kr. In research, wear can be measured in engines fitted with radioactive piston rings; reactions of petroleum, which can be labeled with several isotopes, can be studied; paint film permeability can be measured with tritiated water; and corrosion of metals in salt water labeled with ^{36}Cl can be studied. In environmental studies, silt movement in rivers and sewage pollution of natural waters can be determined.

Tracers may be introduced by adding a homogeneously miscible radioactive material; by adsorbing a radioisotope on the surface; by activation of the material itself; by mechanically inserting a radioactive substance, e.g., a copper wire in a wood chip; and even by labeling another material and using it instead of one that is difficult to label satisfactorily, e.g., a glass fiber for a cellulose one.

Instances of application of tracer methods are virtually unlimited, but they have been developed for troubleshooting or for efficiency studies rather than for routine use. That their potential usefulness may be recognized in practice is the objective of institutes that have been organized in various countries, e.g., Eurisotop in the Euratom community, for collection and dissemination of information on industrial applications of radioisotopes. The increasing availability of sources for activation analysis may increase the use of this technique, which formerly could be used only where a large reactor or neutron accelerator was available.

Massive Irradiation

Several massive-irradiation facilities for production of ethyl bromide and polyethylene, for sterilization of medical supplies and wool, for inhibiting potato sprouting, and for other uses existed at the time of the survey, and others were planned. Economic benefits are difficult to estimate because direct savings result mainly for the customer. Since the survey a number of facilities have been erected, and in a few years much experience with massive-irradiation techniques will have been accumulated.

With regard to the use of ^{60}Co vs. accelerators, at the time of the survey,* for a particular application, the costs per kilowatt-hour equivalent for a gamma source appeared to be twice those for an accelerator. However, the report states: "There are, nevertheless, cases where much more efficient use could be made of gamma radiation; furthermore, the trend is towards decreases in the price of ^{60}Co. Hence, one can foresee that before long a company having several choices of radiation sources will choose a gamma source for economic reasons."

Miscellaneous

A few applications that could not be grouped in the other categories may represent only a small fraction of the total savings but are mentioned because of their potential interest.

Tritium, ^{14}C, ^{90}Sr, ^{147}Pm, and ^{85}Kr are widely used to replace radium in luminous paints for watch and clock dials. The principle used here — emission of light by a solid material, or "phosphor," on exposure to radiation — is used to make nuclear light sources that can be seen for considerable distances. Suggested applications for these sources include safety markers in aircraft, buoys, instrument panel illumination, color identification of equipment used in darkness, and signaling equipment for life rafts. Radioisotopes are already used in railway signal lamps, which compete economically with battery-operated lamps. Because of the long half-life required, only ^{3}H and ^{85}Kr are practical.

The practicality of nuclear batteries has been demonstrated in the U. S. Systems for Nuclear Auxiliary Power (SNAP) program, with ^{90}Sr and ^{238}Pu for satellites.

Sealed radioisotope sources are used for calibration purposes, for example, by X-ray-film producers.

An unusual application that might have great economic importance was reported by one company that grows synthetic quartz crystals. With a nuclear source on one side of the autoclave in

*See *Isotopes and Radiation Technology*, 2(4): 321-328 (Summary 1965), for later information, which indicates that ^{60}Co irradiation may be a factor of 2 to 3 cheaper than accelerator irradiation. Recent reductions in ^{60}Co prices may further decrease isotope irradiation costs.

which the crystals were growing and an X-ray film on the other, the process was watched; bad runs were reduced 25% with an annual saving of $35,000.

USAEC Isotopes and Radiation Development Research and Its Industrial Impact

The AEC's isotopes development program, administered by the Division of Isotopes Development, is directed toward (1) encouraging development of beneficial applications of radioisotopes and radiation technology, particularly those designed to meet problems of urgent public interest; (2) accelerating the potential contribution of radioisotopes and radiation applications to the national economy and welfare; and (3) contributing to world development in the peaceful uses of atomic energy. The objectives are being achieved through a multiphase research and development program.

RADIATION PASTEURIZATION OF FOOD

Research on radiation processing of food is carried out jointly with the Department of the Army. The Army program is concerned with sterilization of food for long-term storage without refrigeration and the AEC program with pasteurization of perishable foods to extend their refrigerated shelf life. Preservation and acceptability factors, packaging, source and facility design, and wholesomeness (with cognizance by AEC's Division of Biology and Medicine) are being studied.

Industry's interest in radiation processing of food has increased notably since the U. S. Food and Drug Administration cleared radiation-sterilized bacon for human consumption in February 1963. Radiation processing of wheat and wheat products and the use of 5-Mev electrons for sterilizing bacon have also been approved. Petitions are pending or planned for sprout inhibition in potatoes; for shelf-life extension of oranges, lemons, peaches, nectarines, carrots, shrimp, haddock, flounder, and cod; and for toxicity clearance for flexible packaging material.*

*Packaging materials approved recently include cellophanes, glassine paper, wax-coated paperboard, polyethylene and polypropylene films, Saran films, and vegetable parchments (*Chemical Week*, p. 27, July 10, 1965).

Current emphasis is on protocommercial facilities intended to prove out laboratory data on a near-commercial scale and to aid in determining the economics. Included in this category are mobile or transportable units, bulk-grain irradiators, and a marine-products development irradiator. The AEC is also actively exploring prospects for establishing a semiproduction plant in Hawaii. Irradiation for quarantine control has shown very encouraging initial results.

PROCESS-RADIATION DEVELOPMENT

The U. S. Government is attempting to provide a productive, balanced effort on radiation processing in concert with that of private industry. An estimated 120 kw of installed radiation power is now being used in the United States for commercial radiation purposes, the major part of which at present involves machine radiation sources. Research is being done on exploratory processing and advanced development systems and on radiation engineering.

Current commercial processes, although still relatively limited in number and dollar value, have shown that continued research on and development of process radiation by both government and industry will broaden the range of productive radiation applications and thus contribute to the national economy and welfare. The program recognizes the appropriate limits to AEC-sponsored research on radiation processing and the beginning of private industry's commercial responsibilities. Thus the AEC's process-radiation development is not directed primarily toward specific product development, but rather toward providing a foundation of radiation technology, not otherwise available, which is basic and of general applicability to commercial development by private industry. Three examples of advanced development projects that have important industrial interest are (1) wood–plastic products, (2) semiconductors by neutron transmutation doping, and (3) ethylene polymerization and copolymerization.

RADIOISOTOPE TECHNOLOGY DEVELOPMENT

Singularly, isotopes represent the greatest potential from atomic energy for doing work of benefit to mankind whether it be economic gain, technological progress, or public-health improvement. Although the basic principles of isotope use have been known for about 25 years, and

important progress has been made in certain areas of use, their broad application is relatively unexplored. Therefore the purpose of the isotopes-technology development activity is to create a broad base of new and improved technology so that use of isotopes in industry, agriculture, medicine, and research can be extended and accelerated. Work is being done in the areas of isotope measurement systems, advanced methods technology, isotope analytical technology, low-level-tracer technology, and applications safety testing.

Of 46 active technical projects conducted under contract with 35 organizations, four representative examples are: (1) development of ^{85}Kr as a universal tracer, (2) application of large-volume detectors to radioisotope process control, (3) use of radioisotope techniques in the canning industry to help protect wholesomeness of food, and (4) industrial use of nuclear techniques for coal analysis.

RADIOISOTOPE PRODUCTION AND SEPARATION TECHNOLOGY

In certain respects the AEC's work on radioisotopes and fission-product production and materials development is one of its most important activities. All its radioisotopes and radiation-technology development effort must ultimately be based on a continuing availability of suitable radioactive materials in adequate quantities and at a reasonable price. Fission, neutron, and cyclotron products; sealed-source safety testing; and advanced technology are the areas of study.

The market for privately produced basic radioisotopes, modified isotope products, and devices in 1962* was $26 million to $32 million:

Sales of privately produced basic radioisotope product forms	>$0.5 million
Sales of labeled organic compounds	$3–4 million
Growth rate	10–15%
Sales of radioactive pharmaceuticals	$5 million
Growth rate	25%
Sales of teletherapy units (seven U. S. manufacturers produced these devices)	$2–3 million
Radiographic equipment and commercial irradiation services	$15–20 million

*This information was brought up to date in a speech by E. E. Fowler, Division of Isotopes Development, which will be published in the next issue of *Isotopes and Radiation Technology*.

ISOTOPIC POWER AND HEAT SOURCES DEVELOPMENT

One of the most exciting new areas of radioisotopes application in recent years is the recovery of thermal energy from radioactive decay; this energy is liberated at a known rate and for a time that is characteristic of the isotope. This has given rise to the development of small nuclear power supplies that can be used in locations and environments where more conventional sources of electric power are unsatisfactory. The incredibly large quantities of suitable radioactive materials that will be required to satisfy isotopic-power needs have posed a severe challenge to the AEC's materials development and production activity. The program includes fuel-forms development (fission products, ^{238}Pu, ^{210}Po, ^{242}Cm, ^{244}Cm, and other neutron products) and study of thermal systems (space propulsion systems, manned space life-support systems, navigation aids, and component heaters).

Currently, isotopic-power fuels are being produced by contractors at AEC production and development sites. It is the policy of the AEC, however, to draw on private industry, to the maximum extent possible, for future production of isotopic-power materials. For example, the Commission has recently invited expressions of interest from industry concerning participation in a large-scale fission-product-recovery program† at the Hanford Works near Richland, Wash. The program is designed to recover fission products from wastes generated in the plutonium-production reactors at Hanford. The Commission is hopeful‡ that private industry will construct and operate the new facilities needed to process and encapsulate the fission products for production and marketing on a completely commercial basis. It is expected that such facilities will cost approximately $9 million to construct. The current schedule calls for the Hanford Fission Products Plant to begin routine operation sometime during 1968.

ANALYSIS AND APPLICATIONS

This field includes study of transfer and use of isotopes and radiation-technology information

†Described in *Isotopes and Radiation Technology*, 2(2): 99–123 (Winter 1964–1965); 3(1): 79 (Fall 1965).

‡The contract has now been let [*Isotopes and Radiation Technology*, 2(3): 316 (Spring 1965); 3(1): 79 (Fall 1965)].

Table V-12 ^{60}Co STERILIZATION FACILITIES FOR MEDICAL SUPPLIES

User	Location	Status	Maximum design capacity (current or proposed initial load), curies
Ethicon, Ltd.	Edinburgh, Scotland	Operational	150,000
Johnson Ethical Plastics, Ltd.	Slough, Buckinghamshire, England	Operational	300,000
Ethicon, Inc.	Somerville, N. J.	Operational	750,000
Ethicon, Inc.	San Angelo, Tex.	Operational	750,000
Ethicon Sutures, Ltd.	Peterborough, Canada	Operational	100,000
Ethicon G.m.b.H.	Hamburg, W. Germany	Being built	750,000
Swann-Morton	Sheffield, England	Operational	10,000
Gillette Industries	Reading, England	Operational	300,000
Gamma Sterilization, Pty, Ltd.	Dandenong, Victoria, Australia	Operational*	3,000,000?
U.K.A.E.A.	Wantage, England	Operational*	500,000?
Newfield Products Ltd.	Mont St. Hilaire, Quebec, Canada	Operational*	600,000
Braun Co.	Melsungen, W. Germany	Operational?	600,000
Tasman Vaccine Laboratory	Upper Hutt, New Zealand	Contemplated since 1963	100–300,000?
Roehr Products Co., Inc.	Deland, Fla.?	Contemplated since 1961	500,000
U.K. Military		Proposed	
USSR	Russia		
Conservatome	Lyon, France	Operational*	70,000?
	Japan	Operational*	

*Part-time basis for medical supplies.

in government, science, and industry. [Establishment of an Isotopes Information Center at ORNL and issuing of a quarterly review, *Isotopes and Radiation Technology*, of information of interest to AEC contractors are included in this field.]

Addendum

Since the IAEA survey was made, the number of massive-irradiation facilities has increased considerably. The facilities for sterilizing medical supplies are listed in Table V-12 as an indication of the current status of this development.

(Martha Gerrard)

Reference

1. *Radioisotope Applications in Industry*, International Atomic Energy Agency, Vienna, 1963 (STI/PUB/70).

Section

VI

Isotopes and Radiation Technology

Miscellaneous Items of Interest

Conferences

Military–Industry Conference on Food Preservation*

A military–industry conference on "Food Preservation and Packaging Techniques" was held under the auspices of the U. S. Army Natick Laboratories and Research and Development Associates, Inc., on Oct. 26–27, 1965, at Natick, Mass. More than 250 persons attended. The first day's sessions are briefly summarized here. Emphasis on the second day was on packaging and is not covered.

Introductory remarks were made by R. L. Hall (McCormick & Co., Inc., Baltimore), by Lt. Gen. W. Vaughan, Col. C. T. Riordan, and F. P. Mehrlich (U. S. Army Natick Laboratories), and by A. I. Totten, Jr., and G. A. Crapple (Research and Development Associates).

In the opening session, N. Raica, Jr. (Fitzsimons General Hospital, Denver) reviewed wholesomeness studies on irradiated food. Wholesomeness tests on sterilized food are essentially complete, but those on pasteurized food are still in progress. Nutrition, microbiology, and induction of toxic materials in sterilized foods are also being studied. Army tests have shown that food sterilized with doses of up to 5.6 Mrads of about 10-Mev electrons is wholesome.

A. Brynjolfsson (U. S. Army Natick Laboratories) stated that standardization is an important factor in electron irradiation of foods. A brief comparison of meat sterilization by machine and radioisotope sources indicates that the action of the two radiations is similar and costs are about the same when the radioisotope sells for $0.25 per curie. He discussed in some detail "spur" formation by electrons.

E. C. Maxie (University of California, Davis) noted that fruits and vegetables "didn't need to rot to go to pot"—handling is a critical factor in preservation of these items. Sterilizing doses of radiation are not suitable for fruits and vegetables, and the range of 200 to 300 krads seems to be feasible for a number of them. He mentioned successful inhibition of ripening in potatoes, processing of onions and asparagus, control of rot in peaches, and disinfestation of a number of products. Radiation has been found unsuitable for processing green Bartlett pears (some have not ripened in 3 years) or green lemons (they ripen fast on the outside but not on the inside). Irradiation preservation of bananas holds considerable promise for commercial application. Some of Maxie's work on strawberries was discussed in *Isotopes and Radiation Technology*, 2(1): 50-54 (Fall 1964).

J. Slavin (Bureau of Commercial Fisheries, Gloucester, Mass.) believes that the two most significant developments in marine-products irradiation in the past year have been (1) optimism replacing doubt about its prospects and (2) start-up of operation of the Marine Products Development Irradiator. More work needs to be done at higher and lower temperatures, and the combination of thermal and irradiation processes should be considered. Many data have been collected on the microbiology of irradiated marine products, but more information is needed on the chemistry of irradiated fish. Also needed are efforts in consumer education; technical assistance to industry; acceptance by consumers, and approval by the U. S. Food and Drug Administra-

*Proceedings will be published in *Activities Report*, Vol. 17, No. 2. Copies may be obtained for $2.00 from: Executive Secretary, Research and Development Associates, Inc., U. S. Army Natick Laboratories, Natick, Mass. 01762.

tion (FDA), of a number of products; and studies on cost, distribution, storage, and shipping. Servicemen are expected to participate in acceptance tests.

H. Ketchum (U. S. Department of Commerce) summarized a recent study done at the Department of Commerce on the potential economic impact of commercial radiation preservation of food [*Isotopes and Radiation Technology*, 3(1): 34-42 (Fall 1965)]. He concluded that the final impact of radiation preservation of foods will depend on the realities of costs, revenues, and profits—all of which reflect the identifiable benefits and consumer satisfactions within a competitive complex.

G. Dietz [U. S. Atomic Energy Commission (AEC)] stated that the AEC's concern was how best to encourage commercialization of radiation preservation of foods. He reviewed the sources used—^{137}Cs, ^{60}Co, and particle and linear accelerators—and emphasized that AEC was not trying to sell isotopes, but a process. He summarized the AEC "family of irradiators" [*Isotopes and Radiation Technology*, 3(1): 30 (Fall 1965)] and mentioned a proposed meat-sterilization pilot plant that was discussed by industry and government representatives on Sept. 24, 1965, at the U. S. Department of Commerce. A cooperative venture by the U. S. Army, AEC, and industry, this plant would sterilize 1 million pounds of meat a year. Industry would construct and operate the plant, AEC would supply design and ^{60}Co, and the Army would buy some of the product and contact other armed services to buy the rest.*

S. A. Goldblith (Massachusetts Institute of Technology) questioned present microbiological safeguards in food preservation. He thinks that these may be unrealistic and that some food canners actually do not live up to the standards expected of processors of radiation-preserved foods. The radiation processor should choose the safest dose for a given product.

C. K. Wadsworth (U. S. Army Natick Laboratories) summarized Army irradiation of beef, chicken, pork, and shrimp. After studies of the effects of radiation on these foods under various conditions, low-temperature irradiation was found to be preferable.

E. Wierbicki (U. S. Army Natick Laboratories) summarized the packaging materials approved for radiation preservation by FDA and described methods of testing them. Besides undergoing tests under different physical conditions, these materials are tested for insect and microbial attack. Data from these tests have been helpful in obtaining FDA approval.

Irradiated foods have been served in 10,000 meals on 150 occasions at Fort Lee. In discussing the results of these tests, F. Heiligman (U. S. Army Natick Laboratories) pointed out that acceptance of most irradiated foods has been proved. More tests are planned, and it is expected that 60,000 meals will have been served by the end of 1971. In these tests refrigerated or frozen unirradiated foods are compared with irradiated foods stored at room temperature for from 3 to 13 months.

E. S. Josephson (U. S. Army Natick Laboratories) pointed out that facilities now operating or planned for radiation preservation of foods are not geared to preserving meats and poultry at doses required to achieve "commercial sterility," that is, doses which yield shelf-stable products with extended unrefrigerated storage life. He strongly recommended the construction of a pilot-plant meat irradiator and read the following U. S. Army policy statement:

> The Chief of Research and Development of the Department of the Army has approved participation by the Army in the proposed extension of the National Food Irradiation Program to include a pilot meat and poultry irradiation–sterilization facility with an annual capacity of approximately 1 million pounds. The other staff agencies of the Army concerned with food have fully concurred in this participation. The role of the Army must be subject to FDA approval of the sterilized meat and poultry items for unlimited human consumption and to development of individual products with reasonable acceptability characteristics. Army support will be in the form of purchase of a share of the total production of the facility for a period of approximately 3 years. Army requirements will be about 150,000 lb each year for large-scale

*The Department of Commerce announced on Jan. 14, 1966, that an interagency task force (Department of Commerce, AEC, Army) is studying the proposal and has scheduled meetings with more than 30 firms interested in the possibilities of establishing a pilot-plant meat irradiator as an industry–government project. The task force is headed by E. R. Killiam, Director of the Office of Chemicals and Consumer Products in the Commerce Department's Business and Defense Services Administration (BDSA). The other members are E. S. Josephson, Associate Director for Food Irradiation, U. S. Army Natick Laboratories, and E. E. Fowler, Director, Division of Isotopes Development, AEC. Companies or organizations interested in the proposal may contact BDSA's Office of Chemicals and Consumer Products.

testing purposes. Discussions are under way with the Navy and Air Force to determine their possible requirements.

Table VI-1 shows the scheduled submission of petitions by the Army for certain radiation-sterilized items.

Table VI-1 SCHEDULED SUBMISSION OF PETITIONS FOR RADIATION-STERILIZED ITEMS

Product	Planned time of submission (fiscal year)
Ham; pork	1966
Beef; chicken; shrimp	1967
Hamburger; corned beef; pork sausage; codfish cakes	1968
Turkey; frankfurters; barbecued beef, chicken, and pork	1969
Luncheon meats; lamb; salmon cakes; baked fish; duck	1970

Radioactive Pharmaceuticals Symposium

A symposium on the use of radioactive pharmaceuticals* was held at Oak Ridge Institute of Nuclear Studies (ORINS), Medical Division, in Oak Ridge, Tenn., Nov. 1–4, 1965. Cochairmen were G. A. Andrews and R. M. Kniseley of ORINS and H. N. Wagner, Jr., of Johns Hopkins Medical Institutions. The meeting, attended by some 200 people, was opened by C. S. Shoup of AEC's Research and Development Division, Oak Ridge Operations, and H. D. Bruner of AEC's Division of Biology and Medicine. R. E. Cunningham (AEC's Division of Materials Licensing), Bruner, and J. Hauser (U. S. Department of Health, Education, and Welfare) considered governmental responsibilities and policies with regard to radioactive drugs.

Internal radiation doses and guides to safe use of radiopharmaceuticals were discussed by E. M. Smith (Hospital for Special Surgery, New York) and G. V. LeRoy (University of Chicago), respectively. Selection of the radiopharmaceutical used in a given case should be based on such considerations as the time-course of the preparation in the target and nontarget areas of the body, the optimum time of making measurements, the dose that can be administered, and methods of detection (H. N. Wagner, Jr., and H. Emmons, Johns Hopkins). Both J. C. Charlton (Amersham) and W. H. Briner (National Institutes of Health) discussed radiochemical purity and sterility requirements, and Y. Cohen (Saclay) discussed these purities in relation to their biological behavior. R. J. Bayly (Amersham) considered the role of labeled compounds in medical diagnosis and M. Blau (Roswell Park Memorial Institute), the biochemical basis for specificity of an organ or tissue for a radiopharmaceutical preparation.

Nuclides for medical use are produced in both reactors and cyclotrons [P. S. Baker and J. J. Pinajian, Oak Ridge National Laboratory (ORNL)], but short-lived isotopes for tracer studies are frequently supplied in the form of generators [P. Richards, Brookhaven National Laboratory (BNL)]. Radioactive microspheres, which are available in graded sizes from 10 to 250 μ, are used for both therapy and diagnosis (I. M. Grotenhuis, Minnesota Mining & Manufacturing Co.).

Radioactively labeled drugs have facilitated experimental pharmacological studies such as the search for an active hypotensive metabolite derived from a drug being tested for its antisecretory function (L. J. Roth, University of Chicago). Double labeling is particularly valuable. An autoradiographic method designed specifically for soluble compounds and based on the dry mounting of freeze-dried frozen sections was described. Activation analysis of drugs labeled with stable isotopes was also used for in vivo diagnoses (W. H. Wahl, Union Carbide Corporation, Sterling Forest).

The iodine isotopes ^{121}I, ^{123}I, ^{124}I, ^{125}I, ^{126}I, and ^{128}I were compared with ^{131}I by W. G. Myers (Ohio State University), and iodinated proteins and other iodinated compounds were reviewed by W. M. Hunter (Clinical Endocrinology Research Unit, Edinburgh) and M. Tubis (Veterans' Administration Center, Los Angeles), respectively. J. P. Kriss (Stanford University) discussed the use of bromine and iodine derivatives of certain biochemicals for tracer studies, and R. S. Yalow (Veterans' Administration Hospital, Bronx) discussed the preparation of high-specific-activity ^{131}I-labeled hormones.

*Oak Ridge Institute of Nuclear Studies, *Radioactive Pharmaceuticals*, AEC Symposium Series, No. 6 (CONF-651111), Oak Ridge, Tenn., 1965.

Technetium-99*m* has favorable characteristics for scanning. P. V. Harper (University of Chicago) discussed the pertechnetate ion, ^{99m}Tc carried on colloidal sulfur, a combination of reduced technetium and iron, tagged albumin, and technetium thiocyanate. A method for preparing ^{99m}Tc-labeled albumin was described by H. S. Stern (Johns Hopkins). The 6-hr ^{99m}Tc may be readily obtained from a generator that contains the parent ^{99}Mo (Richards).

Mercurial compounds were discussed by A. Rothstein (University of Rochester). The synthesis and reactions of sulfur compounds differ somewhat from those of selenium (H. G. Mautner, Yale University), but metabolism of ^{75}Se–methionine is being studied (Blau).

Chromium has become established as a trace element, and ^{51}Cr has been used to study, for example, dialysis in the intestine (C. L. Rollinson, University of Maryland). Intravenously injected chromic phosphate can be used to measure circulation in the liver (E. L. Dobson, University of California).

The labeling of cyanocobalamin with radioactive cobalt isotopes was reviewed by C. Rosenblum (Merck, Sharp & Dohme Research Laboratories). G. C. H. Bauer (Cornell Medical College) discussed calcium and strontium isotopes for bone metabolism studies. E. A. Carr, Jr. (University of Michigan) discussed ^{131}Cs for cardiac and tumor diagnosis, and R. L. Hayes (ORINS) discussed gallium isotopes. H. J. Dworkin (University of Michigan) discussed production of ^{18}F.

Other papers presented were on red cell labeling agents (N. I. Berlin, National Institutes of Health), lanthanides (G. C. Kyker, ORINS), and albumin labeled with various radioisotopes (G. V. Taplin, University of California).

Radioactive gases that can be produced by the Hammersmith Hospital (London) cyclotron are $^{15}O_2$, $C^{15}O_2$, $C^{15}O$, $^{11}CO_2$, ^{11}CO, and $^{13}N_2$; uptake and distribution of ^{133}Xe and $^{13}N_2$ have been simulated with an analog computer (C. M. E. Matthews, Medical Research Council, London). Thin-layer and gas–liquid chromatographic procedures for radioassay of lipids were reviewed by F. Snyder (ORINS).

The radiopharmaceutical industry was the subject of a panel discussion. H. J. Glenn (Abbott Laboratories), M. E. Volk, P. Numerof (E. R. Squibb & Sons), and W. R. Konneker (Nuclear Consultants Corp.) discussed various aspects of industrial supplying of radioactive drugs.

Session chairmen, in addition to Andrews, Kniseley, Kyker, Shoup, Wagner, and Yalow, were A. F. Rupp (ORNL), R. E. Counsell (University of Michigan), and G. Gleason (ORINS).

Mössbauer Effect Symposium

A symposium on the research and industrial applications of the Mössbauer effect was held in New York, Jan. 25, 1966. This conference, which was the second on this subject, was cosponsored by the New England Nuclear Corp. and the Technical Measurement Corp. and was attended by about 175 scientists and engineers interested in the field. The emphasis was on applications of the Mössbauer effect; in the first conference the main emphasis was on techniques and methodology. At the conference it was announced that the proceedings of the first symposium are now available and that the sponsors plan to publish the proceedings of this second conference as rapidly as possible.

The meeting was introduced by I. Gruverman of New England Nuclear Corp. and Lee Grodzins of the Massachusetts Institute of Technology. Abstracts of the papers presented are given below.

Some Applications of the Study of Mössbauer Fractions, Rudi H. Nussbaum (Portland State College). The Mössbauer fraction, $f = \exp(-\langle x^2\rangle/\lambda^2)$, is directly related to the binding of an emitting or absorbing atom in a host lattice. It is often of particular interest if this atom is an impurity. Measurements of f can be applied to the following studies: (1) anisotropic binding of atoms on surfaces or in sites of reduced symmetry inside a host crystal; (2) comparison of host–host with host–impurity force constants and the test of models of crystal lattice dynamics; (3) diffusion mechanisms and the microphysics of phase transitions; and (4) transient nonequilibrium conditions that may be produced by energetic events preceding the Mössbauer emission.

Electric Field Gradients in Iron Compounds, R. W. Grant (North American Aviation Science Center). Investigators have used the Mössbauer effect to determine quadrupole coupling constants in a variety of iron compounds, and some system in the observed values has been noted. The Mössbauer effect permits the rather unique possibility of determining the sign of the electric-field gradient, and the several methods that have been applied for this purpose are discussed. The values (including the sign) determined for the quadrupole coupling constants cast considerable doubt that a general correlation exists between this quantity and the isomeric shift.

Mössbauer Effect Hyperfine Structure in the Presence of Relaxation and Crystal Field Effects, H. H. Wickman (Bell Telephone Laboratories, Inc.). A

simplified introduction to relaxation effects in Mössbauer spectra based on the modified Bloch rate equations is presented. Computed spectra are compared with experimental spectra involving iron or dysprosium compounds. Relaxation effects in the presence and absence of an external magnetic field are illustrated by work from the literature on ferric ammonium sulfate, ferric acetyl acetonate, and certain other iron–organic systems. Hyperfine fields in a number of paramagnetic dysprosium salts are discussed. It is shown that application of an external magnetic field to a paramagnet can markedly affect the Mössbauer spectra and hyperfine fields of the ion. These effects are not related to relaxation but arise from mixing of crystal-field states by the hyperfine interaction and/or the external field. These results are illustrated by Mössbauer experiments with iron-doped Al_2O_3.

Localized Moments of Dilute Fe in Nonmagnetic Host Metals, R. D. Taylor (Los Alamos Scientific Laboratory). The Mössbauer effect of ^{57}Fe has proved useful in studying the magnetic effects of a dilute paramagnetic impurity in a variety of host metals. The hyperfine field, H_{eff}, is studied as a function of temperature and applied field, H. The internal field, generally defined as $H_i = H_{eff} - H$, is interpreted in terms of a modified free-spin behavior, giving rise to a determination of the magnetic moment and spin characteristic of the impurity–host system. The results are compared with localized moments deduced from susceptibility measurements.

Measurement of Magnetization Distributions, W. L. Trousdale (Wesleyan University). In situations where the magnetic atoms of a magnetic material do not all experience the same environment, the Mössbauer method provides a way of determining the local magnetization distribution through the distribution of hyperfine fields. Such situations arise, for example, in substitutionally random ferromagnetic alloys where nuclear magnetic resonance measurements may be difficult. Some methods of calculating magnetic-field distributions are discussed with applications to the palladium–iron system.

Determination of Coordination Numbers for Iron Chemistry, J. J. Spijkerman and F. C. Ruegg (National Bureau of Standards). In Mössbauer spectroscopy the perturbations of the ground and excited states of the nucleus resulting from its chemical environment are reflected in the differences of the chemical shift and the quadrupole splitting. The magnitude of the splitting can be calculated from the *d* electron wave functions. The range of this splitting is calculated for the iron compounds where the iron atom is in an octahedrally symmetric field. Observed deviations indicate a lower symmetry, and the splitting obtained for the M–Fe–EDTA complexes suggests sevenfold coordination.

Mössbauer Analysis of Iron in Stone Meteorites, E. L. Sprenkel-Segel (Argonne National Laboratory) and S. S. Hanna (Stanford University). The absorption of resonance gamma rays by ^{57}Fe has been used in the determination of iron in stone meteorites. By comparison with absorption patterns of terrestrial reference minerals, the meteoritic compounds may be identified and the relative amounts of iron in each mineral determined. Typical minerals analyzed are olivine and pyroxene (ferromagnesian silicates), troilite (a sulfide), and kamacite (an iron–nickel alloy). Iron determination is important in both the classification of meteorites and the investigation of their preterrestrial history. Important inferences concerning the evolution of meteorites can be made from the relative proportions of oxidized and reduced iron in stone meteorites.

Mössbauer Effect in Hemoglobin, George Lang (Carnegie Institute of Technology). A number of hemoglobin compounds have been investigated by means of Mössbauer absorption by the heme iron. In addition to its obvious biological importance, heme is of physical interest because the dilution of paramagnetic ions gives rise to relaxation times sufficiently long that magnetic hyperfine effects are observable. The ferrous compounds with the ligands CO and O_2 and reduced hemoglobin all show simple quadrupole spectra, while in HbNO the unpaired electron gives rise to a magnetic hyperfine interaction. The ferric compounds with CN^-, NO_3^-, OH^-, H_2O, and F^- also exhibit magnetically split spectra, the effect persisting to high temperature only in the fluoride compound. Some progress has been made in interpreting the results and correlating them with the electron spin resonance and susceptibility measurements.

Mössbauer Studies of Rare Earth Intermetallic Compounds, I. Nowik (Bell Telephone Laboratories, Inc.). Compounds of the form AB, AB_2, AB_4, and AB_5, where A is dysprosium or europium and B is any iron, palladium, or platinum transition metal, were investigated by the Mössbauer technique at helium-range temperatures. The effective magnetic field acting on the dysprosium nuclei in all these compounds was found equal, within a few percent, to the expected free-ion value. Only in the compounds with iron and cobalt was the value of H_{eff} higher by 5 to 15% than the free-ion value. The isomeric shifts of dysprosium compounds relative to dysprosium oxide show certain regularities that are still not fully understood. For AB_2 type compounds the isomeric shifts are +1.7 mm/sec for the iron-group compounds, +1.1 mm/sec for the palladium group, and +0.4 mm/sec for the platinum group. Measurements of the intermetallic europium compounds with the use of the 22-kev transition in ^{151}Eu and the 97-kev transition in ^{153}Eu do not show the existence of any anomalous isomeric-shift ratios. An experimental correlation was found between the effective magnetic fields and the isomeric shifts in divalent europium compounds. A simple atomic model is in agreement with this correlation. From the Mössbauer studies some chemical (valency) and magnetic (ordering temperatures) properties of the europium intermetallic compounds were derived.

Mössbauer Effect of ^{119}Sn in Palladium-Rich Tin–Palladium Solid Solutions and the Intermetallic Pd_3Sn, D. K. Snediker (National Bureau of Standards). A study of the palladium-rich end of the palladium–tin alloy system was conducted using the Mössbauer effect of ^{119}Sn to more completely characterize the 9 at.% tin in the palladium Mössbauer source reported by Spijkerman and Herber. Alloy absorbers 10 mils thick, ranging in composition from 2 to 18 at.% tin, and the intermetallics Pd_3Sn and Pd_2Sn were studied with the 9% tin–palladium source. The percentage effect showed a distinct maximum at 13 to 14% tin, corresponding to the resistivity maximum

reported by Knight and Rhys. This resistivity anomaly has been attributed to the filling of the *d* conduction band in the alloy. The Mössbauer spectra of alloys with greater than 10 at.% tin show a distinct doublet nature, the two components having chemical shifts similar to Pd_3Sn and the palladium–tin solid solution. Temperature studies indicate that the doublet is due to two different tin sites and not to quadrupole splitting.

Investigations of Ultrathin Iron Films, C. E. Violet (University of California, Lawrence Radiation Laboratory). Mossbauer resonance spectra of iron films with average thicknesses between 1.2 and 120 A in the temperature range 298–823°K indicate that the hyperfine field, the Curie temperature, and the quadrupole coupling change abruptly for an average film thickness of about 2 lattice parameters. The hyperfine field begins to decrease from bulk value at about 15 A, with a maximum decrease of about 4% of bulk value at 6 A.

Some Applications of Superconducting Magnets, Juergen Heberle (Clark University). Superconducting magnets can generate fields in excess of 100 kgauss. Such field intensities produce observable Zeeman splittings in a number of nuclides that exhibit the Mössbauer effect. When the spectra of sources or absorbers inside solenoids are observed along the direction of the field, the individual Zeeman components are circularly polarized. These features were used in experiments on internal magnetic fields, the Meissner effect, and nuclear magnetic moments. Future applications are expected in studies of E2/M1 mixing amplitudes and unresolved quadrupole interactions. Single-line sources of circularly polarized gamma rays may be feasible.

The proceedings of the first symposium, held in January 1965, are available from Plenum Press, Inc., New York, for $12.50. They contain the following papers:

Mössbauer Spectroscopy: Some Recent Applications to Chemical Problems, Rolfe H. Herber
Review of Advances in Physics, S. L. Ruby
Application of the Mössbauer Effect to Biological Systems, U. Gonser and R. W. Grant
Feedback in Electromechanical Drive Systems, E. Kankeleit
Cam-Driven, Constant-Acceleration Mössbauer Spectrometer, Alan J. Bearden, M. G. Hauser, and P.L. Mattern
Constant-Velocity Mossbauer Drive Systems, P. Flinn
Measurements by Scattering Techniques, I, J. K. Major
Measurements by Scattering Techniques, II, P. Debrunner
f Measurements with Black Absorbers, J. G. Dash
Standardization of the Differential Chemical Shift for ^{57}Fe, J. J. Spijkerman, F. C. Ruegg, and J. R. DeVoe
Computation of Mössbauer Spectra, J. R. Gabriel
Superconducting Magnets—Applications to the Mössbauer Effect, Paul P. Craig
Mössbauer Techniques Using High-Field Water-Cooled Solenoids, Norman A. Blum
Cryostats for Mössbauer Experiments, Michael Kalvius
High-Pressure Techniques, R. Ingalls

AEC Activities

Oak Ridge National Laboratory Ships Record Number of Curies in 1965

S. R. Sapirie, Manager of the AEC's Oak Ridge Operations, said that ORNL made 8484 shipments of radioisotopes in 1965 for a record high total of 1,280,701 curies of activity. This figure more than doubled the old record of 628,352 curies established in 1964. In the 19-year history of ORNL's radioisotope program, more than 202,000 shipments, representing about 4,250,000 curies, have been made.

Also in 1965 the Laboratory completed its largest single commercial order for a radioisotope. In April 173,520 curies of ^{137}Cs was sent to France for use in a mobile irradiation facility in Seine. This was also the largest amount of cesium ever to be loaded in a single container for commerical shipment.

The largest amount of ^{60}Co ever to be shipped in a single container went to American Novawood Corporation, Lynchburg, Va., in December to be used in a pilot-plant production facility. Gamma rays from the 62,640 curies will polymerize a wood–plastic combination to transform a cheap wood base such as pine into a very hard lumber expected to rival mahogany in beauty.

Since private industry has taken over much of the routine production and distribution of many radioisotopes, the number of shipments was down from last year. Commission policy is to withdraw from routine production of regularly used radioisotopes in favor of industry (see the item "Nineteen More . . . Withdrawn . . ." below) to encourage the growth of an independent nuclear industry in the United States.

E. E. Fowler Named Director of Division of Isotopes Development

After serving as Acting Director of the AEC's Division of Isotopes Development (DID) since June 1964, E. E. Fowler has been appointed DID Director, according to R. E. Hollingsworth, General Manager of the AEC. Fowler succeeds Paul C. Aebersold who retired from government service in October 1965. With nearly 18 years of federal government service in the atomic energy field (~10 years of which were spent at Oak Ridge), Mr. Fowler is well qualified to guide AEC's programs in

radioisotope production and separation technology, isotopic power and heat-source development, and radiation application technology.

Agreement States Receive Simplified Regulations on Export and Import of Radioisotopes

Licensees of states that have regulatory agreements with the AEC no longer have to obtain specific licenses before exporting radioisotopes to other countries in the Free World. Similarly, certain forms and quantities of radioisotopes and source materials can now be imported under an AEC general license by persons licensed in these states. Persons in other states will still have to obtain specific licenses from the AEC for export of radioisotopes and import of radioisotopes and source material. The changes became effective Dec. 10, 1965, for Arkansas, California, Florida, Kansas, Kentucky, Mississippi, New York, North Carolina, Oregon, Tennessee, and Texas.

Sandia Laboratory To Provide Technical Administration for Space Projects

The AEC has chosen Sandia Laboratory, Albuquerque, N. Mex., to provide technical direction for the expanding space isotope power program. The Laboratory will report to the Division of Space Nuclear Systems, which has overall responsibility for management of the space isotope power program.

Sandia's new responsibilities will include technical review of component and system designs, hardware, and testing; preparation of specifications; establishment of quality-control standards; reliability evaluation; and development-program review and planning. The Laboratory will also conduct in-house technology development to support the system development activities, point the direction for future development work, and maintain its competence in the isotopic power field.

The AEC Albuquerque Operations Office will administer the industrial contracts for which the Laboratory will provide the technical direction. Full transfer to Albuquerque and Sandia of contract administration and technical direction, respectively, from the AEC New York Operations Office will require about 6 months.

Availability of Isotopes and Services

Cobalt-60 Prices Plunge as Specific Activity Rises

For the first time since late 1963, the AEC has lowered prices of ^{60}Co. The new price schedule, which took effect Dec. 18, 1965, establishes a new category of 1 to 15 curies/g and increases the maximum limit on specific

Table VI-2 NEW AND OLD PRICE SCHEDULES FOR COBALT-60

New Schedule

Specific activity, curies/g	Single-order quantity, curies	Price per curie
Up to 15	100,000 or more	$0.40
15 and to 30	100,000 or more	0.50
30 and to 45	100,000 to 250,000	0.65
	250,001 to 500,000	0.60
	500,001 or more	0.55

Special Provision. The Commission will not supply ^{60}Co (1) in quantities less than 100,000 curies or (2) in specific activities over 30 curies/g in other than BNL strip form, except—in each case—when domestic commercial producers cannot meet requirements under reasonable conditions of time, quality of product, or price. Under these conditions, prices are:

Specific activity, curies/g	Price per curie	Single-order quantity, curies	Discount, %
Up to 30	$2.00	Up to 5,000	None
30 and to 40	3.00	5,000 and to 25,000	15
40 and to 55	4.00	25,000 and to 100,000	30
55 and to 70	5.25		
70 and to 85	5.65		
85 and to 100	5.85		
100 and to 115	6.05		
115 and to 130	6.20		
130 and to 145	6.40		
145 and to 160	6.60		
160 and to 175	6.80		
175 and to 190	7.00		
190 and to 205	7.20		

Old Schedule

Specific activity, curies/g	Price per curie	Single-order quantity, curies	Discount, %
1–25	$2.00	0–5000	None
26–40	3.00	5,001–25,000	15
41–55	4.00	25,001–100,000	30
56–70	5.25	^{60}Co with a specific activity of 30 curies/g or less is priced at $0.50 per curie for custom orders greater than 100,000 curies.	
71–85	5.65		
86–100	5.85		
101–115	6.05		
116–130	6.20		
131–145	6.40		
146–160	6.60		
161–175	6.80		
176–190	7.00		
191–205	7.20		

activity supplied by the Commission from 30 to 45 curies/g. The new low-activity category will cost $0.40 per curie on minimum orders of 100,000 curies. In the range from 30 to 45 curies/g, the cobalt will be supplied only in the form of "BNL" strips, and prices will range from $0.55 to $0.65 per curie, depending on how much is purchased. The new and old price schedules are shown in Table VI-2.

Under its policy established in 1961, the AEC will supply ^{60}Co only when commercial producers cannot meet reasonable requirements of time, quality of product, or price. No quantities less than 100,000 curies will be provided unless the material is not reasonably available from commercial sources.

Nineteen More Radioisotopes May Be Withdrawn from Routine Sale and Production

A proposal has been received by the AEC to withdraw from routine sale and production 19 radioisotopes currently available through AEC's Oak Ridge National Laboratory. These are ^{124}Sb, ^{76}As, ^{77}As, ^{82}Br, ^{109}Cd, ^{115}Cd, ^{115m}Cd, ^{64}Cu, ^{198}Au, ^{199}Au, ^{140}La, ^{197}Hg, ^{203}Hg, ^{99}Mo, ^{32}P, ^{42}K, ^{110m}Ag, ^{24}Na, ^{35}S (sulfate). Used principally in research and medical diagnosis and therapy, the 19 radioisotopes accounted for $231,485 of sales from ORNL during fiscal year 1965 in 4012 shipments totaling 114,334 mc.

As in past withdrawals the AEC will continue to meet requirements to the extent that the purchaser certifies in writing that he requires material of a technical quality which is not commercially available.

ORNL Stable-Isotope Inventory Grows

Recent additions of highly enriched stable isotopes to the ORNL inventory include small quantities of 95.4% ^{123}Te and about 64% ^{196}Hg and multigram quantities of 98.9% ^{28}Si, 93.6% ^{57}Fe, 99.66% ^{64}Zn, and 99.4% ^{128}Te.

Standard Gamma-Ray Spectra Available from ORNL

The ORNL Isotopes Development Center announces the availability of a collection of gamma-ray spectra for 42 nuclides (see Table VI-3). Statistically treated spectra were obtained from absolutely assayed sources with a standard gamma-ray spectrometer system [10 cm distance from a 3- by 3-in. NaI(Tl) detector]. One may use the standard gamma-ray spectra for both qualitative and quantitative analysis for these nuclides without the need for radioactivity standards. This form of analysis may be accomplished by insertion of the digital data from punched-paper tape into a multichannel analyzer memory for a direct efficiency determination. A report describing the standard spectra collection and its use will be published in *Nucleonics*.

Table VI-3 NUCLIDES COMPRISING THE STANDARD LIBRARY

Nuclide	Standard number	Nuclide	Standard number
^{7}Be	17	^{95}Nb	24
^{22}Na	3	^{99}Mo*	40
^{22}Na	10	^{106}Ru*	16
^{24}Na	7	^{110}Ag	33
^{42}K	19	^{124}Sb	34
^{46}Sc	5	^{125}Sb*	39
^{47}Sc	43	^{130}I	32
^{51}Cr	6	^{131}I	14
^{54}Mn	8	^{134}Cs	15
^{56}Co	38	^{137}Cs*	4
^{56}Mn	11	^{140}Ba*	22
^{59}Fe	13	^{140}La	23
^{60}Co	2	^{141}Ce	29
^{64}Cu	27	^{144}Ce*	41
^{65}Zn	35	^{147}Nd	21
^{72}Ga	20	^{153}Sm	42
^{75}Se	28	^{181}Hf	30
^{76}As	37	^{182}Ta	31
^{82}Br	26	^{191}Os	36
^{85}Sr	18	^{192}Ir	25
^{86}Rb	12	^{198}Au	9

*Indicates equilibrium with radioactive daughter.

The gamma-ray spectra are available in several forms: punched-paper tape (several different formats), graphical displays, and typewriter readout of the digital data. To obtain copies of any or all of the standard spectra, contact Isotopes Development Center, Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830.

General

Isotopes Information Center at ORNL Now Officially Open

The Isotopes Information Center (IIC), operated by ORNL for the AEC, was formally announced in March 1966. The IIC was established "to collect, evaluate, and disseminate worldwide information on production and uses

of radioisotopes in industry and research." The Center also maintains a current awareness of uses of isotopes in biology, medicine, agriculture, and education.

The IIC publishes *Isotopes and Radiation Technology*, issues state-of-the-art reviews, and answers specific requests for information, in addition to its other activities. A full report on the IIC will be published in the next issue of this Review. For further information contact: P. S. Baker, Director, Isotopes Information Center, Oak Ridge National Laboratory, Post Office Box X, Oak Ridge, Tenn. 37830.

Radiation Chemistry Data Center Established at Notre Dame

As part of the National Standard Reference Data System, a radiation chemistry data center has been set up at the University of Notre Dame, Indiana. Jointly sponsored by the National Bureau of Standards and AEC, the new facility will compile and evaluate data on chemical reactions brought about by ionizing radiations. In addition, the facility will serve as an international focal point for the collection, storage, evaluation, and dissemination of radiation chemistry data. Information will be evaluated by scientists now working in the field of radiation chemistry throughout the world.

The center is under the direction of Professor Milton Burton, Director of the University's radiation laboratory, and is located in the radiation research building built on the campus by the AEC in 1963. Initial efforts will be concerned with the compilation, critical evaluation, and interpretation of available data relating to reactions induced by ionizing radiations. Types of data to be compiled include chemical-reaction yields, effects on physical properties, and specific rates of elementary processes.

Selected Current Literature

W. S. Lyon, Jr. (Ed.), *Guide to Activation Analysis*, D. Van Nostrand Company, Inc., Princeton, N. J., 1964 (186 + xix pp., $5.95); reviewed by Andrew Kramer.

Prepared under the auspices of the U. S. Atomic Energy Commission, this book on activation analysis was written by members of the Nuclear-Radiochemical Group of the Analytical Chemistry Division of Oak Ridge National Laboratory.

It need scarcely be mentioned here that activation analysis has become an extremely important peacetime use of atomic energy. It has taken its place in the laboratory as a versatile and sensitive tool and has opened new horizons in analysis of elements throughout all branches of science and industry.

Guide to Activation Analysis was written to help chemist–analysts through the relatively unfamiliar territory of nuclear physics, reactor technology, and radiation detection and counting. It examines the unique things about the field—the equipment used to do the activation, the activating particles, and the techniques and equipment needed to measure the induced activity. It points out potential pitfalls, indicates when activation analysis can be used advantageously, and, equally important, when it cannot. It shows by precept what interferences may be encountered and how they may be minimized.

To a large extent, the book is intended for the newcomer in the field who wants to get started in the technique without too much personal assistance. It comprises a concentration of the results of experiences gathered from the performance of over 300,000 nuclear analyses. The text presents the principles and methods of activation analysis and proceeds to examine the applied technology. Finally, the principles that have been developed are *applied* to the solution of representative activation analysis problems; calculations in these problems are shown in detail.

The two appendices contain an invaluable store of information. Appendix A lists the theoretical calculated sensitivity for each element and typical concentrations found in a variety of matrices. Appendix B provides information on safe-handling techniques for radioisotopes and on licensing requirements.

The idea for a *Guide to Activation Analysis* was first proposed by the AEC's Advisory Committee on Isotope and Radiation Development. The editor, William S. Lyon, Jr., is supervisor of the Nuclear-Radiochemical Group in the Analytical Chemistry Division at ORNL. He has been prominent in the field of radiochemistry and nuclear measurements

since 1944. ORNL contributors to the volume include authorities in all of the several activation techniques and associated technology. Their background reflects a diversity of relevant experiences. Two were formerly on the training staff at Oak Ridge Associated Universities; two came from Brookhaven; and one was formerly a staff member of both the Argentine Atomic Energy Commission and the Chalk River facility in Canada.

P. S. Baker, D. A. Fuccillo, Jr., M. W. Gerrard, and R. H. Lafferty, Jr., *Radioisotopes in Industry*, U. S. Atomic Energy Commission, November 1965 (50 pp.).

This booklet, one of the USAEC's Understanding the Atom series, was prepared by members of the Isotopes Information Center at ORNL. It gives a broad, popular survey of ways that radioisotopes are used in industry. Single copies may be obtained free from USAEC, P. O. Box 62, Oak Ridge, Tenn. 37830.

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Franz Buschke (Ed.), *Progress in Radiation Therapy*, Vol. III, Grune & Stratton, New York, 1965 (242 pp., 370 refs., $14.50).

Of the 13 chapters in this book, all but two are documented. High-energy-electron and supervoltage therapy techniques are discussed, and X-ray, radium, and cobalt-beam treatment of bone and mammary cancer and radiation therapy for children are evaluated. A section on radiation therapy education will be of interest to persons planning medical school curriculums.

A. Charlesby (Ed.), *Radiation Sources*, The Macmillan Company, New York, 1964 (268 + vii pp., $9.00).

The subjects covered (by 11 authors) are nuclear reactors as radiation sources, isotopic power sources, Van de Graaff accelerators, cylindrical electrostatic generators, resonant-transformer electron-beam generators, linear accelerators for radiation processing, personnel protection in irradiation facilities, and neutron flux and energy measurements. Seven of the eight chapters are documented. Author and subject indexes are included.

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E. Gradsztajn and R. Bernas, A Simple Method for Preparing ^{7}Be Through the Action of Fast Protons on Water, in *Radioisotopes in the Physical Sciences and Industry*, Conference Proceedings, Copenhagen, 1960, International Atomic Energy Agency, Vienna,

*CFSTI is Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151 (formerly Office of Technical Services).

Vol. 2, pp. 453-454 (in French); ORNL-tr-172, translated by J. J. Pinajian and Martha Gerrard.

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J. K. Lancaster, A Review of Radio-Active Tracer Applications in Friction, Lubrication and Wear, British Report RAE-TN-CPM-64, March 1964.

John H. Lawrence (Ed.), *Progress in Atomic Medicine*, Vol. 1, Grune & Stratton, New York, 1965 (233 pp., $9.75).

This first volume in a planned series illustrates the use of radioactive isotopes in medical research, diagnosis, and therapy. Among the subjects covered are whole-body counters, activation analysis of biological materials, scanning, use of short-lived isotopes and positron emitters, and therapy with both external sources and internally administered isotopes. Individual, well-documented chapters were prepared by Anger, Berlin, Cohn, Collins, Fateeva, Gottschalk, Lawrence, Siri, Snyder, Söremark, Ter-Pogossian, and Tobias.

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